4.0 CONSTITUENTS IN RECYCLED URANIUM

4.1 INFORMATION SEARCH AND DATA SOURCES

The project team searched a variety of data collections and libraries at ETTP and other ORR locations to identify and retrieve analytical data. Much of the data found was located in the K-1034 retired records vault, the Environmental Restoration (ER) Information Center at ETTP, or the Information Resource Center. This information was supplemented by data gathered from Bechtel Jacobs organizations at ETTP (e.g., Radcon and Analytical Laboratories). Major data sources consulted and analyzed included:

- ORGDP historical site reports, including quarterly plant reports and engineering progress reports
- ORGDP historical technical and experimental research reports
- ORGDP reports describing operations and production processes
- Plant records, including employment, health physics, and environmental monitoring and materials release records
- ORGDP production records
- ORGDP analytical laboratory records
- Correspondence between shippers and receivers
- Historical DOE and contractor reports addressing RU
- More recent (i.e., post-1990) health physics reports and databases
- More recent environmental survey and safety basis reports (e.g., Basis for Interim Operation documentation, characterization reports, and hazard screenings)
- Environmental reports submitted to state and federal agencies

In addition to consulting the ORGDP analytical laboratory records, the team found it necessary to glean analytical data from a wide variety of sources, including the ORGDP historical quarterly reports, technical reports, environmental reports, and health physics reports. Correspondence between shippers and receivers also provided a record of comparisons of sets of analytical data (the first set developed by the site shipping RU and the second by the site receiving the material). In addition, analytical data has been compared and shared with other appropriate DOE sites.

For some areas that presented gaps in data that could not at present be filled by research, the project team developed estimates for quantities of RU and/or constituents. These estimates are based on extrapolations from actual data and represent (1) application of known data from similar material and/or circumstances or (2) application of known data from a specific time period over a longer or a shorter period of time. All such estimates and their bases are specifically identified in this report.

The approach used in searching for and collecting data useful to the project team's purpose was suitably comprehensive in terms of targeting the broad range of likely sources and locations of data. However, because of time and resource limitations, the

Site Team could not absolutely verify that all relevant and useable analytical data and records were identified and reviewed.

As a result of the brief but intensive search, the project team determined that a significant amount of information exists to address the scope and objectives established for this phase of the RU project. Further, results of this current effort have extended previous evaluations and have, in some instances, served to confirm earlier work. With respect to constituent analysis, a reasonable quantity of data was found and evaluated.

4.1.1 K-25 Analytical Laboratory Information System

Beginning in 1985, K-25 Site analytical laboratory information was captured in the K-25 Analytical Information Laboratory System (KANLIS). Analytical records prior to 1985 did not fall under the 75-year rule and were shipped to Atlanta for long-term storage. Whether the records were retained beyond five years was not confirmed. Previous experience indicates that the probability of retrieving records from the Atlanta repository is remote. Therefore, the Site Team focused its efforts on data available from ORGDP. Hard copy records of analytical data since 1985 (in KANLIS) are stored either in the K-1034 Retired Records Vault or at the Y-12 Plant. A database maintained by the analytical laboratory uses sample number and QA number to determine the physical location of the hard copy record. The team determined that it was feasible to use KANLIS as a potential source of quantitative TRU data.[1]

The current KANLIS was queried to identify all records with laboratory analyses for Pu, Np and ^{99}Tc. This query identified approximately 700,000 records, which were scanned for building numbers and descriptions of interest to narrow the data set to approximately 70,000 records. Another scan identified 150 records that appeared to be the most relevant for the project. Table 4.1-1 presents a sample of these 150 records.

Table 4.1-1. Example of Records of Potential Interest Identified from KANLIS

Sample Number and Date Completed	Constituent	Result	Units	Description
850502-079	Tc	<0.005	µg/gU	ORGDP Tails
May 1985	Pu	<1	dpm/gU	ORGDP Tails
	Np	5	dpm/gU	ORGDP Tails
850723-118	Tc	3.65 E2	µg/gSample	Inc Ash 1420
August 1985	Pu	2	dpm/gSample	Inc Ash 1420
	Np	10	dpm/gSample	Inc Ash 1420
850723-119	Tc	5.16 E1	µg/gSample	Inc Ash 1420
August 1985	Pu	11	dpm/gSample	Inc Ash 1420
	Np	7	dpm/gSample	Inc Ash 1420
850723-120	Tc	2.62 E1	µg/gSample	Inc Ash 1420
August 1985	Pu	23	dpm/gSample	Inc Ash 1420
	Np	43	dpm/gSample	Inc Ash 1420
850924-056	Tc	0.040	µg/ml	Cylinder Wash
October 1985	Pu	11.75	dpm/g	Cylinder Wash
	Np	14.46	dpm/g	Cylinder Wash

[1] KANLIS Analytical Laboratory Data, March 1985 to current (received April 2000).

Unfortunately, these samples identified contained no QA number, and the location of the hard copy record could therefore not be determined through the database.

4.2 ANALYTICAL LABORATORIES

The ORGDP Analytical Laboratories are located in Buildings 1004-A, -B, -C, and -D. The laboratories have been in operation at the site since 1944.[2] In the earliest years of the plant, the physical and organizational sections of the Laboratory Division were the Conditioning Section, the Sampling Section, the Special Analysis Section, the Spectrometer Section, and the Uranium Analysis Section. Analyses performed and procedures for the activities within the Laboratory Division sections are described in a 772-page *K-25 Works Laboratory Manual* dated December 1952.[3] Some specific examples of information in the manual of interest to this report included:

- The Industrial Hygiene Group within the Special Analysis Section performed urinalysis using a procedure that involved evaporation, dilution, and electroplating uranium, with the results of the analysis reported in alpha counts/min/100 ml of urine.
- A measurement control program was maintained for the Mass Spectrometer Section (within the Special Analysis Section) to closely track current measures of the precision of all types of routine analyses to enable adequate steps to be taken to keep the precision of all analyses within the desired control limits.
- The Uranium Analysis Section maintained a measurement control program and issued a monthly *Quality Control Report* based on data obtained by measuring control batch materials.
- The Counting Group (within the Uranium Analysis Section) followed detailed procedures for alpha counting in urine, alpha activity in air and water, and beta-gamma activity in water, residues, and recovered UO_3.

At the time this manual was issued, no analysis methods were listed for ^{99}Tc. It is noted that alpha counting instrumentation prior to the early 1960s did not discriminate the energy level of the alpha particle counted and thus did not discriminate the isotopic source of the alpha activity detected. The source of alpha activity so detected could have been from uranium, Pu, Np, or any other isotope decaying via alpha particle emission.

Prior to the development of instrumentation (alpha spectroscopy) for discriminating among the energy levels of alpha particles, some urinalysis was performed specifically for Pu and Np. The urinalysis was accomplished by separation of the elements by chemical means before the sample was alpha counted. The limited amount of data available from these analyses may suggest that urinalysis was performed only for specific cases in which transuranic exposure was suspected.

[2] W. B. Humes, K-25 Plant Superintendent, to C.D.W. Thornton, U.S. AEC, correspondence, January 21, 1949.
[3] *K-25 Works Laboratory Manual*, K-990, Carbide and Carbon Chemicals Company, January 2, 1953.

More recent documentation shows that detailed practices and procedures continued to be used and refined for Analytical Laboratory activities.[4] During this period (circa 1973), the laboratory functions included such services as sampling, sub-sampling, determination of chemical purity and specific impurities, and radiochemical and isotopic analyses. Laboratory functions of particular included:

- Uranium samples representing shipments to and from other AEC installations, uranium processors, and licensees were sub-sampled and distributed for various specification analyses.
- Purge cells and/or other off-stream equipment were sampled and analyzed for uranium. Samples were also removed for mass spectral analyses.
- The abundance of the various uranium isotopes was determined for UF_6 samples. These samples came from specified points in the cascade; from cylinders received, stored, or shipped; or from other uranium compounds which were fluorinated to UF_6.
- Radiochemical analyses were performed to quantitatively determine concentrations of various radioisotopes. Appropriate extraction procedures and detection instruments were selected for specific samples. Quantitative alpha, beta, and gamma radioactivity measurements on a variety of samples including air, water, soil, vegetation, and special materials were performed.
- The Special Analysis Section determined uranium content in samples of urine and other biological materials in support of the Industrial Hygiene program for personnel protection. This section also performed analyses of plant effluents, vegetation, and mud samples for purposes of pollution monitoring.

A number of historical reports and documents containing information on analytical and sampling practices were found and reviewed. *AEC Accountability Survey Reports*[5] evidence a practice of on-going, contemporaneous review and evaluation of analytical and sampling practices during the period when RU was received and processed at ORGDP. Examples include:

- "The program whereby station HGE (Hanford) samples each lot of depleted uranium trioxide and sends the sample under separate cover to K-25 for analysis as representative of the lot of material has proven satisfactory. Carbide K-25 has compared their own analysis of Hanford-supplied samples with the K-25 analysis of samples taken from the lots of material on a random basis at K-25. These analyses agree with the expected limits of error of the x-ray photometric method of analysis so that the Hanford-supplied sample is considered as representative of trioxide received by Carbide K-25. The random sampling of lots by Carbide K-25 was continued as a control program."

- "Sampling Methods. The sample exchange program between the K-25 Plant and Hanford and Harshaw are apparently under satisfactory control. An independent

[4] *Nuclear Materials Management Manual*, K-P-4086, Rev. 4, Union Carbide Nuclear Company, Oak Ridge Gaseous Diffusion Plant, 1973.
[5] *AEC Accountability Survey Reports, Reports for the Period October 1947 through May 27, 1953 (U)*, KZ-7801-31.

sample is taken of every fifth lot at K-25 to ascertain that the sample supplied by the shipper is adequate. There is no significant difference between the analyses made at K-25 and those made at Hanford or Harshaw."

- "Uranium trioxide from Harshaw is shipped in 16 drum lots (grossing about 800 pounds per drum) with two lots comprising a shipment of about 12 tons of oxide. A sample taken as the drums are filled is supplied for each lot. These two lot samples are composited at the K-25 Plant and one analysis is made for uranium content and for isotopic ratio. Similar material is received from Hanford in shipments of 12 lots of 8 drums per lot, about 40 tons of material per shipment. Analysis is made on a lot basis in this case also, with the sample being supplied by Hanford. Spot samples are taken at K-25 from every fifth lot."

4.3 HISTORIC STANDARDS/SPECIFICATIONS REGARDING MAXIMUM ALLOWABLE TRANSURANIC AND FISSION PRODUCT CONTENT IN RECYCLED URANIUM

A mutually agreeable and technically sound transuranic and fission product element specification between shipper and receiver for all recycle material shipped to and from all DOE sites handling recycle material was recommended in the Egli Report in 1985.[6] The report stated that such a specification had probably never existed either within or between sites. Although most sites had their own "working" specification, there simply was no understanding and agreement on specifications for recycle material shipped to or from the DOE sites. Having said that, the task force further found that there were *informal* standards or specifications that were used within and between sites. Table 4.3.1 summarizes the various specifications that were (or might have been) used by ORGDP or by organizations supplying or receiving material to or from ORGDP.

Table 4.3-1. Summary of RU Specifications

Specification	Source
Maximum Alpha Activity from all Transuranic Elements: 15,000 dpm	AEC
Hanford UO_3 Product: Pu <10 ppb	Hanford
Maximum Alpha Activity from all Transuranic Elements: 1,500 dpm	DOE – Savannah River
Maximum Alpha Activity from all Transuranic Elements: 3,000 dpm	DOE – Oak Ridge
Pu-239 < 10 ppb	DOE – Oak Ridge and FMPC
Alpha: $\frac{\text{(Activity per gram of Pu + Np +Th) x 700}}{\text{(Nominal Activity of Enriched Uranium)}} \leq 1.0$ **Beta**: $\frac{\text{(Activity of Sample)}}{\text{(Activity of Unirradiated Uranium Standard)}} \leq 1.25$ **Gamma**: Total Fission Product ≤ 0.20 : Ci/g Uranium	Y-12
Transuranic Alpha <5,000 dpm/g U	ICPP

[6] D. Egli, et al., *The Report on the Joint Task Force on Uranium Recycle Materials Processing*, DOE/OR-859, U.S. DOE Oak Ridge Operations, September 1985.

The American Society for Testing and Materials (ASTM) has two specifications for UF_6: one for UF_6 that is intended for feeding an enrichment plant and the other for enriched UF_6 product (up to 5% ^{235}U). These specifications are:

- C787-96 "Standard Specification for Uranium Hexafluoride for Enrichment"
- C996-96 "Standard Specification for Uranium Hexafluoride Enriched to Less Than 5 Percent ^{235}U"

It should be noted that these specifications apply to uranium used as fuel in commercial power reactors exposed to very high levels of burnup compared to production reactors. However, the cooling time for discharged fuel from production reactors was very much less. The fuel "slugs" were sent for reprocessing after cooling for 50 days to allow fission products (^{131}I) to decay. The ASTM specifications for power reactor fuel are based on a cooling period of ten years to accommodate the burnup level of 50,000 megawatt-days per MTU.

Prior to 1966, the permissible concentration of transuranic elements in UF_6 feed was 150 alpha dpm/gU.[7] In December 1966, (31FR16584) the AEC announced a tenfold relaxation of that level. This adjustment was made in anticipation that reactor returns would not average more than three tons per day in the period 1967 through 1975 and that the additional transuranic elements fed to the diffusion plants during that time could be tolerated without significant additional health risks.

The first specifications for UF_6 delivered to or by the AEC were published in *Federal Register*, 23 F.R. 4813, dated June 28, 1958. Federal Register Notice, Volume 32, Number 230, Wednesday, November 29, 1967, which is the genesis of the current UF_6 specifications, consolidated, revised, and superseded all previous notices. The product specifications were minimal—calling for UF_6 content of at least 99.5 wt % UF_6 and containing 0.01 mol % of hydrocarbons, partially substituted halo-hydrocarbons, or chlorocarbons. The feed specifications had limits for gamma and beta activity from fission products and alpha activity from all transuranics. The gamma activity was 20% of the gamma activity of aged natural uranium, and the beta activity was 10%. The alpha activity was 1500 disintergations per minute (dpm) of total uranium. Current ASTM specifications are based on the same gamma and alpha activities. The beta activity has been replaced with a specification on ^{99}Tc.

In October 1988, DOE put into effect a feed specification based on ASTM 787. Finally, in June 1994, ASTM 787 and ASTM 996 were adopted as the specifications for UF_6 feed and product for DOE enrichment plants. Both of the current ASTM specifications provide for feed and product derived from RU. The following appears in both ASTM 787 and ASTM 996:

1. Reprocessed UF_6—any UF_6 made from uranium that has been exposed in a neutron irradiation facility and subsequently chemically separated from the fission products and transuranic isotopes so generated.
2. Discussion—The requirements for Reprocessed UF_6 given in this specification are intended to be typical of reprocessed spent fuel that has achieved burnup

[7] R.W. Levin, *UF_6 Specifications for Feed for Gaseous Diffusion Plants*, K/TL-1092 Rev. 1, October 1, 1981.

levels of up to 50,000 MW days per tonne of uranium in light water reactors and has been cooled for ten years after discharge. It is recognized that different limits would be necessary to accommodate different fuel histories.

ASTM 787 has the following requirements for reprocessed uranium:

1. For Reprocessed UF_6, the gamma radiation from fission products shall not exceed 1.1×10^5 <MeV Bq/kg U (1.1×10^5 MeV/sec kg U).
2. For Reprocessed UF_6, the alpha activity from neptunium (Np) and plutonium (Pu) isotopes may be specified in either of two ways as agreed upon between the parties concerned:
   a) The total alpha activity from Np and Pu in the cylinder shall be limited to 25,000 Bq/kgU (1.5×10^6 disintegrations per minute per kilogram of uranium), or
   b) The volatile alpha activity from Np and Pu in the liquid sample from the shipping container shall be limited to 3,300 Bq/kgU (0.2×10^6 disintegrations per minute per kilogram of uranium).
3. For Reprocessed UF_6 the concentration of ^{99}Tc shall be measured and reported. It shall not exceed 0.500 micrograms per gram of total uranium (µg/gU).
4. Minor isotopes in reprocessed UF_6 shall not exceed the limits given as micrograms per gram total uranium (: g/gU):

^{232}U	0.005
^{234}U	480.0
^{236}U	8400.0

ASTM 996 has the following requirements for gamma and alpha activity:

1. For Enriched Reprocessed UF_6, the gamma radiation from fission products shall not exceed 4.4×10^5 MeV/sec kgU.
2. For Enriched Reprocessed UF_6, the alpha activity from neptunium and plutonium shall be less that 3,300 Bq/kgU (200,000 dpm/kgU).
3. The specification for minor isotopes represent limits obtainable from the enrichment of reprocessed UF_6 feed materials at the corresponding limits of Specification C 787:

   ^{232}U 0.050 µg/gU
   ^{234}U 2000 µg/gU
   ^{99}Tc 5 µg/gU

   Note: Depending upon the demands placed on fuel fabricators and reactor operators, it may be necessary to agree on lower limits.

4.4 ANALYTICAL RESULTS FOR TRU AND FISSION PRODUCTS IN RECYCLED URANIUM MATERIALS RECEIVED AT ORGDP

On an activity basis, the principal radionuclides expected to pass through chemical processing of reactor returns and remain in the RU received are the TRU radionuclides produced in highest abundance and with moderate half-lives: ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, and ^{241}Am. In addition, certain fission and activation products may form volatile compounds in the fluorination process (^{99}Tc, ^{106}Ru, ^{125}Sb, ^{134}Cs, and ^{137}Cs). Some operational data from the 1950s and 1960s indicates the presence of detectable quantities of ^{95}Zr-Nb, ^{106}Ru, and ^{137}Cs in ORGDP materials. However, because ^{95}Zr-Nb, ^{106}Ru, and ^{134}Cs have short half-lives (65 days, 368 days, and 2.1 years, respectively) and RU was last introduced in 1984, they are unlikely to be present in significant quantities today.[8]

From the beginning, the presence of non-uranium constituents in RU receipts and the introduction of these contaminants into the ORGDP facilities and equipment as a result of processing those receipts were recognized. Evidence indicates that RU that was to be shipped to or was received at ORGDP was systematically sampled, with checks performed for TRU and fission products. Records of analytical data for ORGDP RU receipts were found to exist in the Building K-1034 Retired Records Vault. These records consisted of correspondence from the ORGDP Laboratory Division Superintendent reporting results obtained from analysis of material lot samples submitted to ORGDP by Hanford, Harshaw, and Savannah River during the 1952 through 1957 time period. Dates for this set of analytical data were found to correspond to the years that RU was received from Hanford, Harshaw, and Savannah River as determined from MBRs.[9] These analytical results are thus considered as representative of the material received at ORGDP during the same time periods.

The analytical results, as reported in the correspondence found, were compiled and reviewed. Typically, the reported results included weight % ^{235}U, Pu in ppb, total beta activity and total gamma activity, and fission product beta and gamma activity. Beta and gamma activities were reported as a percentage relative to the beta or gamma activity of an equal weight of natural uranium in equilibrium with its daughters.

Analysis for Pu was not always performed. Contemporaneous correspondence indicated that from time to time an understanding existed between shipper and receiver that Pu was not expected to vary from earlier shipments because reprocessing process parameters remained unchanged. One example, which referred to UO_3 from Hanford, stated: "…and the ratio of plutonium produced versus UO_3 shipped has been fairly constant over the past few years and is not expected to change significantly in the near future."[10] In another example, the good agreement between results obtained from separate analyses performed by ORGDP and by Savannah River on the same material is cited as a sufficient basis to discontinue the practice of duplicate ORGDP analysis on

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[8] *Assessment of Accessible Contamination at the K-25 Site Phase 3 Report: Cumulative Analytical Results*, K/HS-570, Oak Ridge K-25 Site, May 1994.

[9] For the mass balance study, receipts were identified as RU based on the site from which the material was received and the assay range of the material, as determined from information contained in Material Balance Reports (MBRs). Historical Forms 741 for the shipments of interest, which could provide actual shipment dates or transfer numbers to directly link the material balance data to the analytical data, were not found.

[10] W.L. Richardson to R.L. Dagley, "Shipment of UO_3," Union Carbide Internal Correspondence, June 22, 1962.

each lot of UO_3 from Savannah River.[11] In the case of material from Harshaw, it appears that a practice of sampling for Pu at a 1 in 4 frequency was adopted for the latter shipments. Similarly, beta and gamma was not always measured; rather, the data suggests that these were measured until it was judged that the material was sufficiently characterized.

Table 4.4-1 provides a summary of the Pu results for receipts of RU from domestic sources.[12] A weighted average was calculated for each source, based on annual averages and annual amounts of material received from each source. Pu data for receipts of RU from Harshaw, Hanford and Savannah River are presented in Fig. 4.4-1, 4.4-2, and 4.4-3, respectively. As can be seen in Figure 4.4-2 for material received from Hanford, Pu analytical results dated between January and April 1953 are significantly higher than for any other period.

Table 4.4-1. Summary of Data on Plutonium in Early ORGDP RU Receipts

Fiscal Year	No. of Lots	No. of Results	Max Pu *(ppb)*	Min Pu *(ppb)*	Avg Pu *(ppb)*	Total U *(kgs)*
Hanford						
1952	81	78	12	1	2.1	99,970
1953	109	92	40	1	13.7	578,249
1954	26	26	4	1	1.4	1,115,345
1955	10	5	2	1	1.2	526,475
Total	226	201			4.5	2,320,039
Harshaw						
1953	148	67	11	1	3.2	1,402,761
1954	7	2	9	1	5.0	299,574
Total	155	69			3.5	1,702,335
Savannah River						
1955	47	47	9	1	3.8	271,949
1956	256	19	8	2	4.6	2,538,844
Total	303	66			4.5	2,810,793

[11] J. C. Barton, Works Laboratory Superintendent, Union Carbide Nuclear Company, to W.H. Emslie, E.I. duPont deNemours and Company, "Analysis of SRO and K-25 samples from Lots 101 through 111," October 11, 1955.

[12] Where Pu results were reported as less than a given limit value, a value equal to 50% of the limit was used (i.e., for a reported result of <2 , the average of the range 0 to 2 was used, or 50% of 2=1).

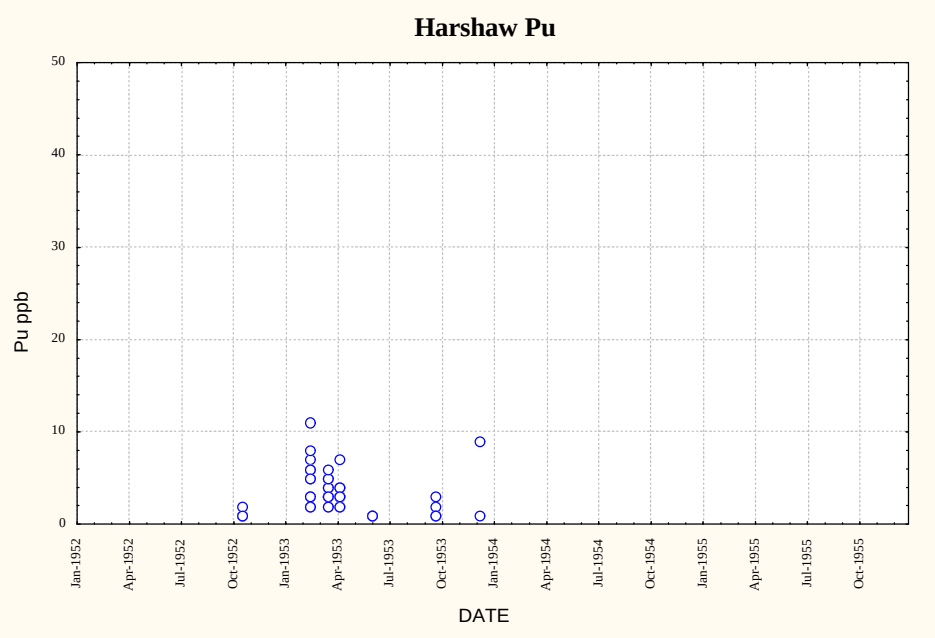

Fig. 4.4-1. Plutonium in ORGDP RU Receipts from Harshaw.

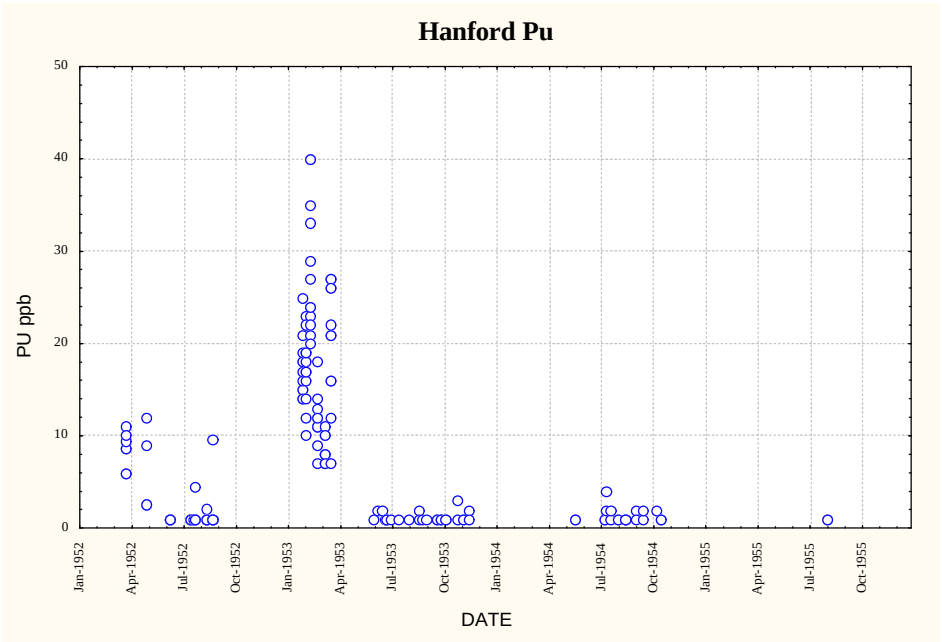

Fig. 4.4-2. Plutonium in ORGDP RU Receipts from Hanford.

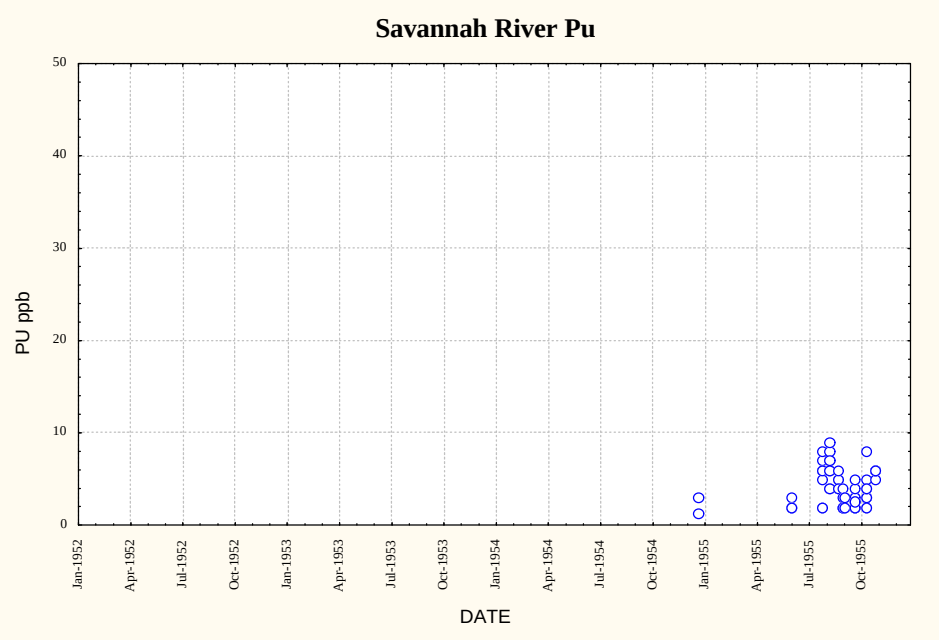

Fig. 4.4-3. Plutonium in ORGDP RU Receipts from Savannah River.

Total gamma activity results are presented in Fig. 4.4-4, 4.4-5, and 4.4-6. Total beta activity results are presented in Fig. 4.4-7, 4.4-8, and 4.4-9. Fission product beta activity results for material from Hanford and Savannah River are presented in Fig. 4.4-10 and 4.4-11 and fission product gamma activity results are shown in Fig. 4.4-12 and 4.4-13. There was no fission product activity data reported in this record set for material from Harshaw.

In addition to domestic sources of RU, ORGDP also received RU from foreign sources. From 1969 to 1988, ORGDP received 1,294 MTU of foreign RU reactor returns. Information was found in a series of reports of natural and reactor return feed analyses.[13, 14, 15, 16] The reports summarized results of sampling and analysis performed at ORGDP for defining adherence to feed specifications. All cylinders of reactor return UF_6 were sampled and analyzed for full specifications. It was reported that during the period of 1969 through 1982, eight cylinders of reactor returns from COGEMA (French) failed to meet specifications (six for transuranic alpha and one each for fission product beta and gamma). No cylinders of foreign reactor returns were reported as exceeding the specifications for TRU and fission products for the period of 1983 through 1986.

[13] W. D. Hedge, *Toll Enrichment Uranium Hexafluoride: Natural and Reactor Return Feed Analyses at ORGDP for CY 1982, Including Summaries for CYS 1969-1982*, K/TL/AT-58, Rev. 1 Addendum 2, Union Carbide Corporation Nuclear Division, April 1983.
[14] W. D. Hedge, *Analyses of ORGDP Toll Enrichment Uranium Hexafluoride for CY 1983*, K/PS-5034, Union Carbide Corporation, March 1984.
[15] W. D. Hedge, *Analyses of ORGDP Toll Enrichment Uranium Hexafluoride for CY 1984*, K/PS-5034, Addendum 1, Union Carbide Corporation, May 1985.
[16] W. D. Hedge, *Analyses of ORGDP Toll Enrichment Uranium Hexafluoride for CY 1985 Through September 1986*, K/PS-5034, Addendum 2, Union Carbide Corporation, January 1987.

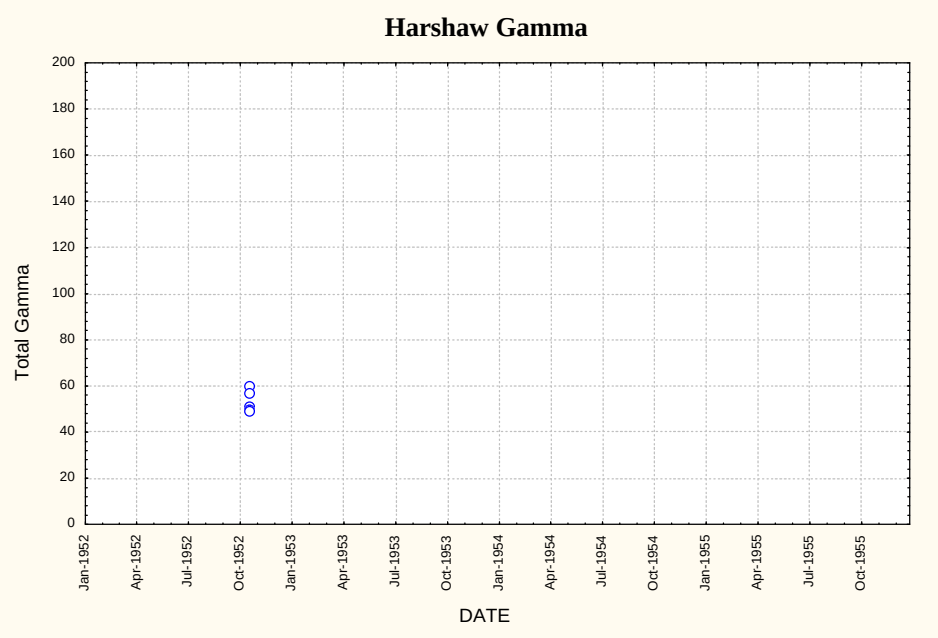

Fig. 4.4-4. Total Gamma Activity for ORGDP RU Receipts from Harshaw.

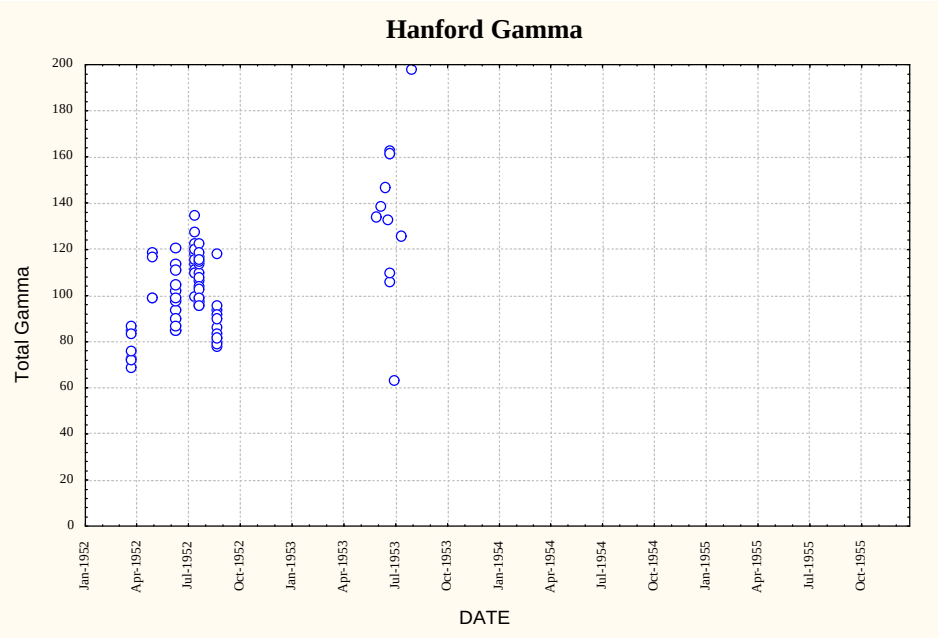

Fig. 4.4-5. Total Gamma Activity for ORGDP RU Receipts from Hanford.

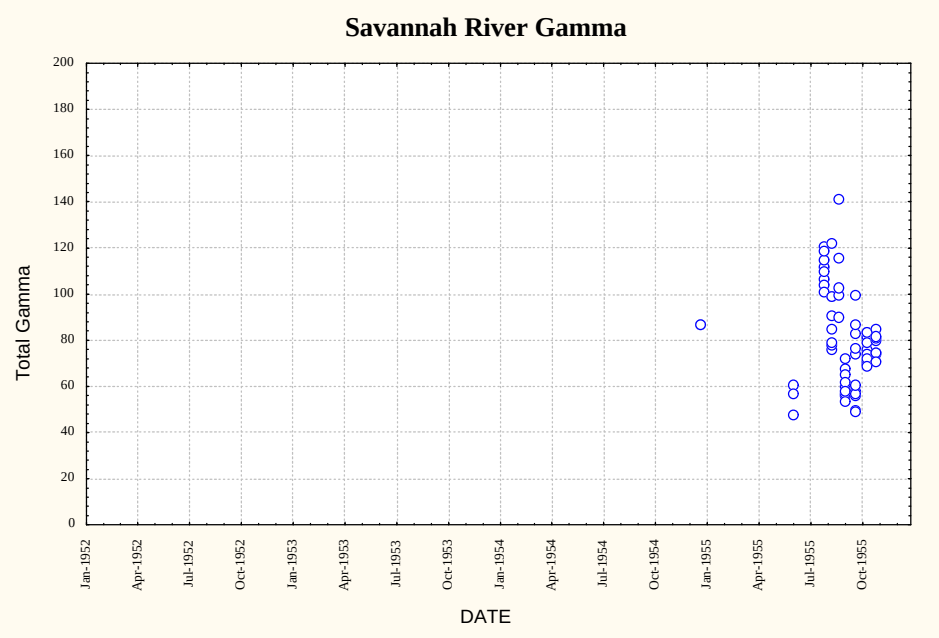

Fig. 4.4-6. Total Gamma Activity for ORGDP RU Receipts from Savannah River.

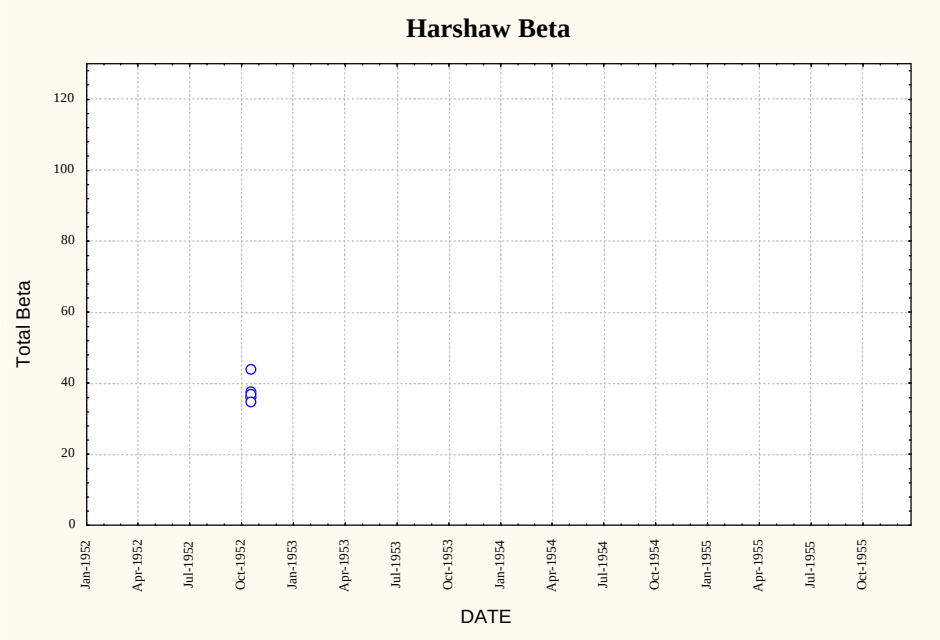

Fig. 4.4-7. Total Beta Activity for ORGDP RU Receipts from Harshaw (% relative to normal uranium standard).

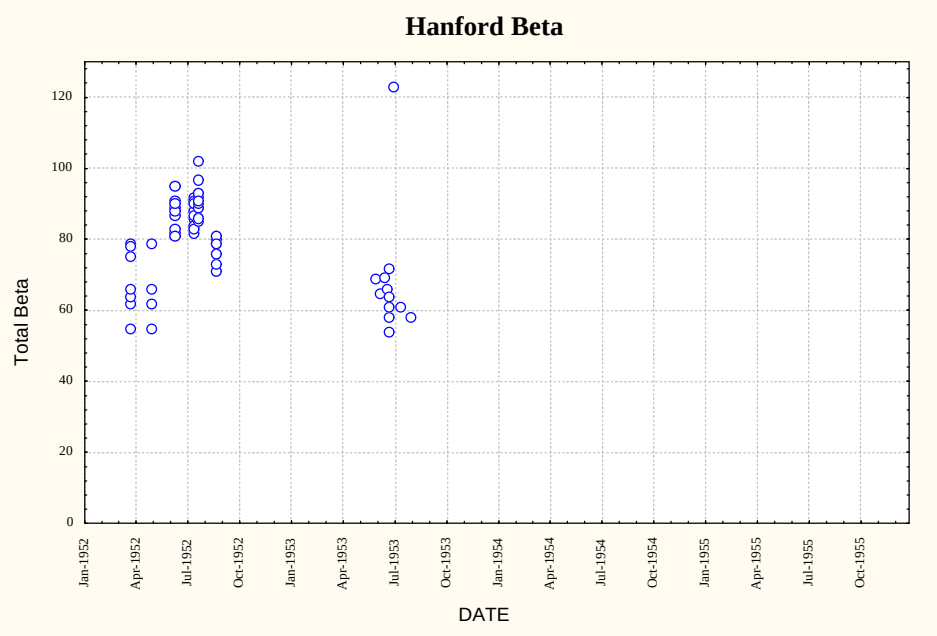

Fig. 4.4-8. Total Beta Activity for ORGDP RU Receipts from Hanford (% relative to normal uranium standard).

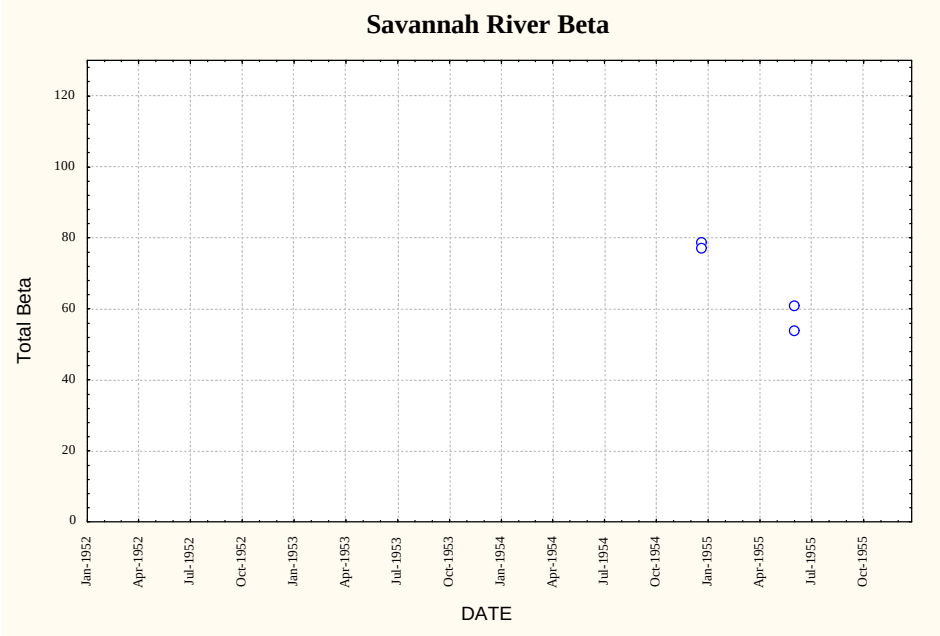

Fig. 4.4-9. Total Beta Activity for ORGDP RU Receipts from Savannah River (% relative to normal uranium standard).

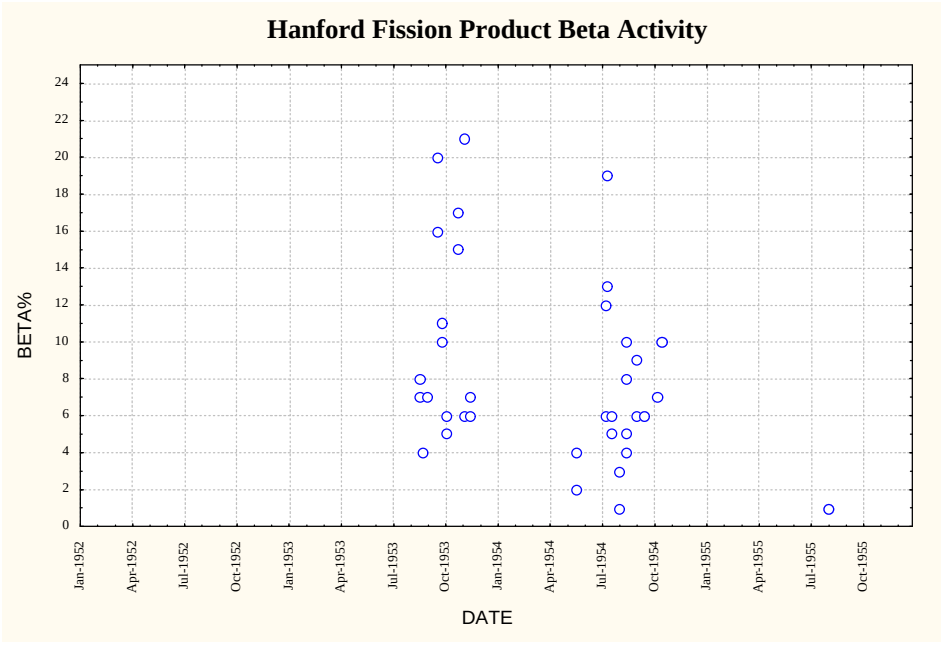

Fig. 4.4-10. Fission Product Beta Activity for ORGDP RU Receipts from Hanford.

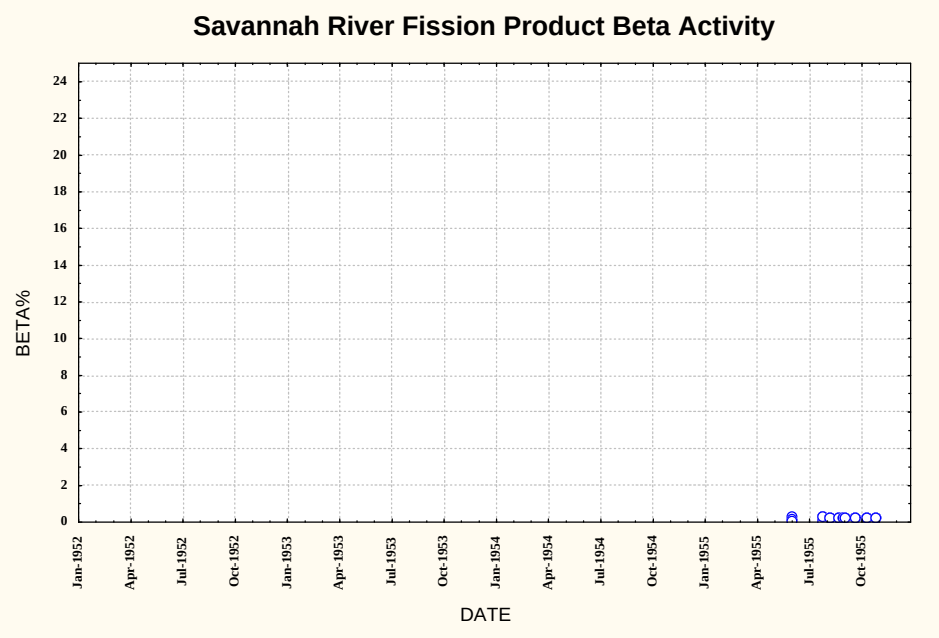

Fig. 4.4-11. Fission Product Beta Activity for ORGDP RU Receipts from Savannah River.

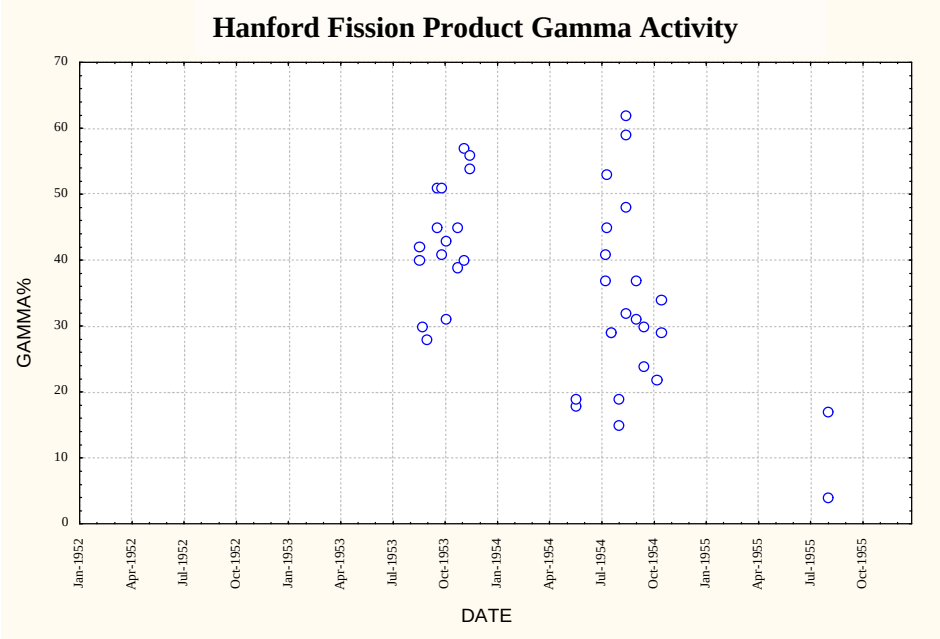

Fig. 4.4-12. Fission Product Gamma Activity for ORGDP RU Receipts from Hanford.

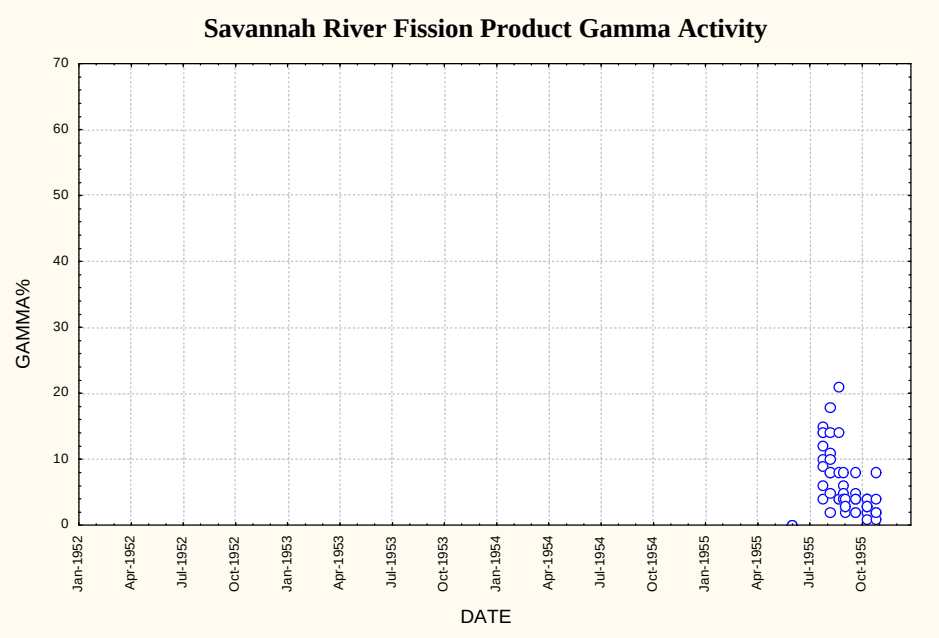

Fig. 4.4-13. Fission Product Gamma Activity for ORGDP RU Receipts from Savannah River.

Table 4.4-2 summarizes, by year, results of analysis for TRU and fission products in material from foreign fuel reprocessors. Transuranic alpha was observed in most reactor return samples.

Table 4.4-2. Analyses of TRU and Fission Products in ORGDP RU Receipts from Foreign Sources

CY	Property	U-235 *(wt%)*	U-236 *(wt%)*	Fission Product Gamma *(% Aged Natural U)*	Fission Product Beta *(% Aged Natural U)*	TRU Alpha *(dpm/gU)*	Tc *(ppm U)*
1969	French (Cogema)	1.62300	0.0390	100.0	13.00	<150.0	
1970	Belgian (Euro-Chem)	0.95830	0.1850	<5.0	<2.00	134.0	
1970	British (BNFL)	1.80500	0.0500	<5.0	<2.00	186.0	
1970	French (Cogema)	1.76700	0.2320	6.2	<2.00	1,323.0	
1972	British (BNFL)	1.91700	0.0710	<5.0	<2.00	386.0	
1972	French (Cogema)	1.51800	0.1580	<5.0	<2.00	180.0	
1973	British (BNFL)	1.37500	0.0450	<5.0	<2.00	140.0	
1973	French (Cogema)	1.97700	0.2420	<5.0	<3.00	748.0	
1974	British (BNFL)	0.64230	0.0110	<5.0	<2.00	170.0	
1974	French (Cogema)	1.50700	0.1760	<5.0	<3.00	250.0	
1975	British (BNFL)	0.83900	0.0120	<5.0	<2.00	42.0	
1976	French (Cogema)	2.01700	0.0110	<5.0	<2.00	<25.0	
1976	British (BNFL)	0.64200	0.0220	<5.0	<2.00	<10.0	
1977	Belgian (Euro-Chem)	1.05270	0.0290	<5.0	<2.00	44.0	
1977	British (BNFL)	2.06400	0.0570	<5.0	<2.00	<25.0	
1978	British (BNFL)	1.06200	0.0510	<5.0	<2.00	5.0	
1978	French (Cogema)	1.04600	0.1520	<5.0	<2.00	30.0	
1979	French (Cogema)	1.02900	0.0240	<5.0	<0.30	18.0	
1979	German	2.01500	0.0280	<5.0	<0.30	<5.0	
1980	French (Cogema)	1.07000	0.2540	6.4	<3.00	42.0	
1980	Russian	2.67800	0.0160	<5.0	<0.30	<5.0	
1981	French (Cogema)	1.01300	0.2390	6.7	0.44	6.3	
1982	French (Cogema)	1.31100	0.2400	<5.0	0.73	6.1	0.041
1983	French (Cogema)	1.03462	0.2835	<0.1	0.20	5.2	0.022
1983	Netherlands (Urenco)	1.96135	0.3180	9.0	0.20	<5.0	0.008
1983	Russian	2.67762	0.0172	<0.1	<5.00	<5.0	<0.000
1984	French (Cogema)	1.18864	0.2918	9.7	2.30	5.7	0.006
1984	Germany	3.09910	0.0023				
1985	French (Cogema)	1.21652	0.3257	6.1	3.00	3.5	0.013

4.5 ANALYTICAL RESULTS FOR TRU AND FISSION PRODUCTS IN RECYCLED URANIUM PROCESS STREAMS AND WASTE STREAMS AT ORGDP

4.5.1 Feed Plant Ash

In a 1957 ORNL paper[17], Lantz and Parker note that a moderate amount of Np was discovered in uranium oxide at ORGDP. Lantz and Parker state that sampling and analysis of various material streams from the PGDP metal recovery plant, which

[17] P. M. Lantz and G. W. Parker, "Investigation of Paducah Ash and Metal Recovery Waste as a Large-Scale Source of Neptunium-237," Oak Ridge National Laboratory, July 1957.

processed all of the waste from the diffusion plant, including ash from the feed plant, was subsequently undertaken. Results indicated that most of the Np was deposited in the nonvolatile ash collected in the feed plant. No neptunium was found in the product uranium or tails, and no samples were available from intermediate stages.

Records from Fernald indicate that two shipments of feed plant ash from PGDP were received at Fernald in 1978 and 1980. Analytical results for samples of the 1980 shipment exist and were provided by Fernald.[18] While the data provided are for ash shipped to Fernald from PGDP, it is reasonable to assume that the constituent nature of the PGDP ash is similar to that produced by ORGDP. ORGDP shipped much of its feed plant ash to PGDP, and some ORGDP ash was directly included in PGDP ash. The ORGDP feed plant also operated in a nearly identical manner as the PGDP feed plant.

Analytical data was provided for 16 samples taken from 16 hoppers containing a total net weight of 40,651 kg of material. The net weight contained by individual hoppers ranged from 1,394 kg to 4,147 kg. Analytical data provided included wt % U, wt % ^{235}U, Np in disintegrations per minute per gram (dpm/g) sample, total Pu in dpm/g sample, and ^{99}Tc in dpm/g sample. The mass fractions of Np, Pu, and ^{99}Tc in parts per billion parts U (ppb U) were calculated using the analytical results and specific activities for the given radionuclides. For Pu, the conversion was performed assuming the Pu was 100% ^{239}Pu. Table 4.5.1-1 summarizes the mass fraction data for Np, Pu, and ^{99}Tc.

Table 4.5.1-1. Summary of Data for Paducah Feed Plant Ash Shipped to Fernald in 1980

	^{99}Tc *(ppb U)*	Np *(ppb U)*	Pu *(ppb U)*
Sample Population	16	16	16
Mean	3,091	6,724	1,262
Median	1,652	4,434	385
Minimum	354	1,173	67
Maximum	11,977	25,287	7,747

Mass fraction data for Np, Pu and ^{99}Tc plotted against wt% ^{235}U are shown in Fig. 4.5-1, 4.5-2 and 4.5-3, respectively. With such a small sample population, meaningful extension of the result using statistical methods is unlikely. However, the data appear to support the inference that a large fraction of the Np in the RU feed partitions to and becomes concentrated in the feed plant ash. Given the average of 1,262 for Pu in ash, and considering an average Pu concentration of 4 ppb in feed to the feed plant, the data in Fig. 4.5-2 supports the conclusion that Pu partitions to and becomes concentrated in the feed plant ash waste stream.

[18] C. W. Lowery, Fernald, facsimile transmittal of "Paducah Feed Plant Ash Received in Hoppers, 1980," April 6, 2000.

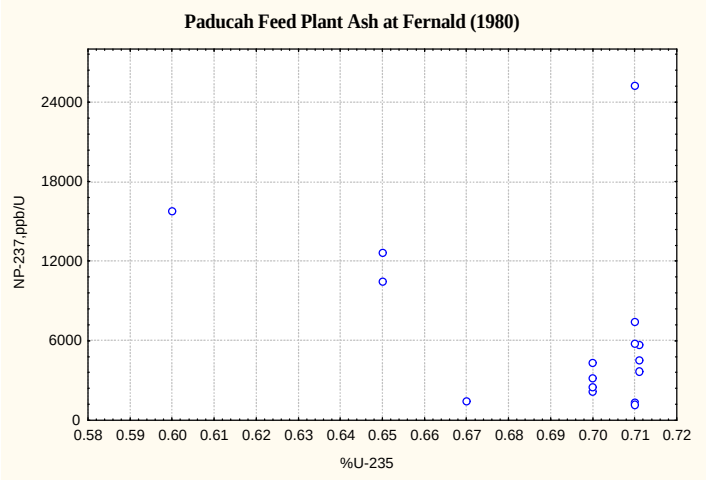

Fig. 4.5-1. Neptunium in PGDP Feed Plant Ash Shipped to Fernald in 1980.

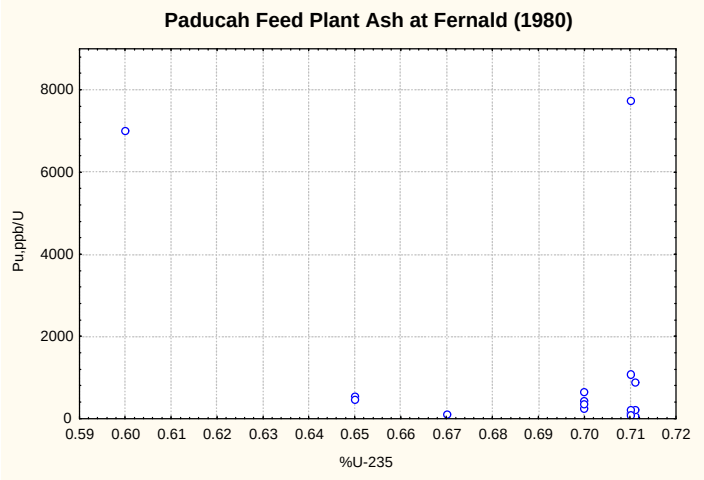

Fig. 4.5-2. Plutonium in PGDP Feed Plant Ash Shipped to Fernald in 1980.

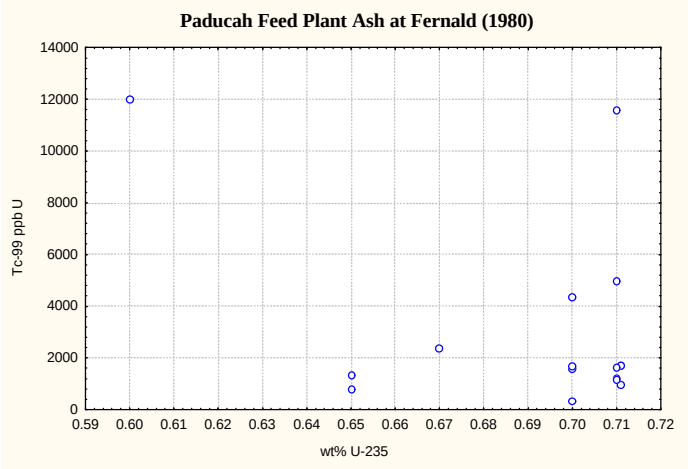

Fig. 4.5-3. Technetium in PGDP Feed Plant Ash Shipped to Fernald in 1980.

Additional data dealing specifically with Pu content was found in a 1953, K-1131 feed plant sampling report.[19] Between 15 and 41 samples were taken of four ash streams and analyzed for Pu (reported in ppb U). The data is summarized in Table 4.5.1-2.

Table 4.5.1-2. Pu in K-1131 Feed Plant

Pu *(ppb U)*	Mean	Min	Max
Flange 1	1,455	13	8,100
Flange 2	905	50	3,900
Flange 3	169	5	1,100
Barrier Filter Powder	769	0	14,800

The flange (1 through 3) samples are believed to be ash streams from the fluorination bed in the feed plant. The barrier filter powder is the ash stream from the vent at the end of the feed plant process. This data also supports the conclusion that Pu in RU processed through the feed plant partitioned to and became concentrated in the feed plant ash.

The data is shown graphically in Fig. 4.5-7. A period of higher Pu results is seen during April and May 1953, which generally corresponds with the period when high Pu results were seen in the Hanford material receipts (i.e., January through March 1953).

[19] A. F. Becher to J. Dykstra, *Monthly Plutonium Report*, Union Carbide Nuclear Company, Internal correspondence, January 1953 through July1961

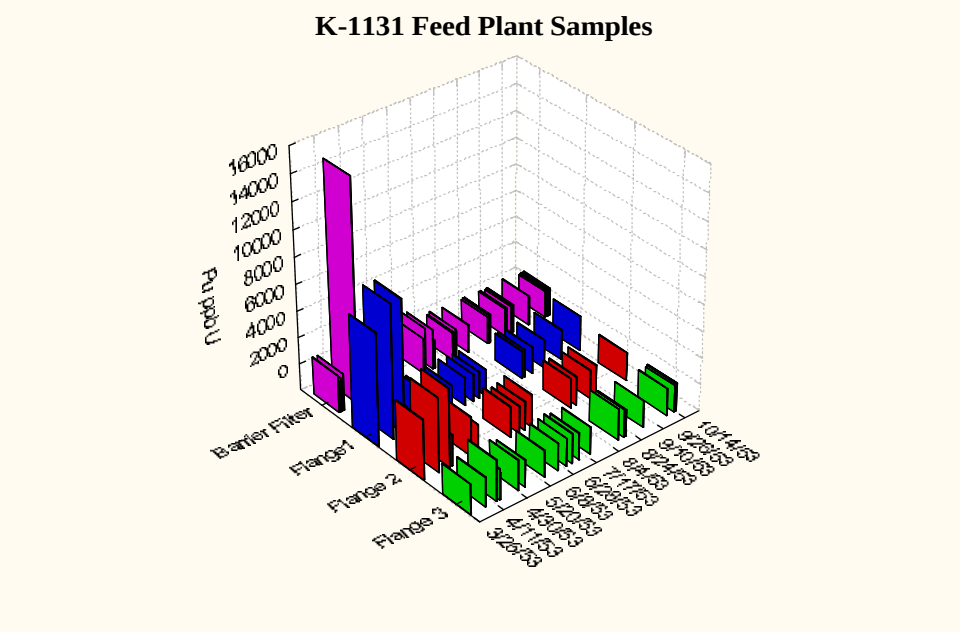

Fig. 4.5-7. Plutonium in K-1131 Feed Plant Samples.

4.5.2 Neptunium in the K-33 Building

Information was found in the *ORGDP Quarterly Report*[20] for the fourth quarter of 1962 regarding a sampling program conducted in the K-33 portion of the cascade. The sampling program was designed to detect the presence of radioactive contaminants other than uranium and its daughter products and to follow the movement of these contaminants through the cascade. Samples of barrier and converter deposits were obtained from converters in the K-33 Building. The report notes that since Np is ten times more harmful than uranium on an alpha basis, more stringent safety precautions are required when the Np concentration is found in excess of the "acceptable limit of 10 μgNp/gU."

The report states that the highest Np to uranium ratio in any deposit obtained from cascade equipment occurred in a gray powder that was observed on or near the converter B-outlet tube sheet. Reported results are shown and summarized in Table 4.5-2.

Table 4.5-2. Results for K-33 Cascade Equipment Deposits

Location	Sample Date	U (g/g)	Np (mg/g)	Np:U (mg/g)
K-902-1.9-6	09/08/61	0.100	3.80	38
K-902-1.8-6	08/30/61	0.084	3.10	37
K-902-2.7-2	12/10/61	0.066	3.10	47
K-902-2.7-3	02/24/61	0.066	3.70	56
K-902-3.3-1	11/14/61	0.150	12.00	80
K-902-3.5-3	08/08/61	0.070	0.80	11
K-902-3.5-6	07/25/61	0.130	1.30	10
K-902-3.9-5	10/25/61	0.076	2.10	28
K-902-3.10-3	10/13/61	0.092	1.60	17
K-902-4.5-4	12/08/61	0.066	4.90	75
K-902-4.5-5	09/22/61	0.059	5.30	90
K-902-4.5-6	09/24/61	0.069	4.70	68
K-902-5.1-1	09/24/61	0.075	15.00	200
K-902-5.8-1	03/07/61	0.150	0.02	0.1
K-902-5.8-4	01/23/62	0.046	9.60	210
K-902-7.7-5	12/29/61	0.030	0.80	27
K-902-7.2-3	10/19/61	0.037	0.30	8
Sample Population		17	17	17
Mean		0.08	4.242	58.9471
Median		0.07	3.1	38
Range		0.12	14.98	209.9
Minimum		0.03	0.02	0.1
Maximum		0.15	15	210

The origin of the powder and the mechanism by which Np was retained in the cascade were documented as "not known at the present time." The gray powder was relatively inaccessible in the converter and would only be encountered when the equipment was entered for inspection or when the converter was being disassembled. Many other samples were documented as having been examined from cascade equipment, including converters, compressors, and piping, but only the gray powder showed the presence of Np in concentrations in excess of the limit of 10 μgNp/gU. As of the writing of the fourth quarter report, samples with Np to uranium ratios in excess of this value were confined to K-33; however, Np may have been present in K-602-2.6.

4.5.3 Technetium Removal Plant Test

Information was found in the *ORGDP Quarterly Report*[21] for the fourth quarter of 1962 concerning a plant test using MgF_2 traps to remove ^{99}Tc from the cascade in Building K-33. Removal of ^{99}Tc from PGDP product UF_6 by sorption on MgF_2 was investigated using traps installed at the K-33 feed point. The report states that ^{99}Tc in the ORGDP cascade feed streams was of concern because it can cause a 1 to 2% loss in barrier permeability. At that time, the value of ^{99}Tc was believed to be approximately

[20] *ORGDP Quarterly Report*, April 1, 1962 through June 30, 1962 (Q4)
[21] *ORGDP Quarterly Report*, April 1, 1962 through June 30, 1962 (Q4)

$90/gm, thus making recovery of the material more desirable. Analytical results from the test showed that most of the ^{99}Tc had been sorbed in the first and second bed sections, with almost none sorbed at the discharge end of the trap.

Table 4.5-3. Impurities Scavenged from PGDP UF_6 by MgF_2

Spectrographic analyses of impurity concentration, ppm			
Bed 1	Bed 2	Bed 3	Bed 4
8000	3000	<20	<20

Total grams sorbed: 12.0 grams
Equivalent reduction in impurity concentration: 1.0 ppm

The amount of ^{99}Tc trapped represents a reduction of 1.0 to 1.4 ppm in the UF_6 processed through the trap. This reduction was consistent with the observation of negligible concentration of ^{99}Tc in the trap outlet and an estimated average value of 1.6 ppm ^{99}Tc in the PGDP product flow. Np concentration on the sorbent suggests its presence in the PGDP product at approximately 0.35 ppb, which was well below the 20 ppb limit of detection at that time.

4.6 ANALYTICAL RESULTS FOR TRU AND FISSION PRODUCTS IN FACILITIES AND EQUIPMENT IN WHICH RECYCLED URANIUM WAS PROCESSED AT ORGDP

Early in ORGDP's operating history, the presence of non-uranium constituents in ORGDP RU receipts and the introduction of non-uranium contaminants into the facilities and equipment as a result of processing RU at ORGDP was recognized. A number of related studies and historical reports have been found to contain information and data useful for this project.

4.6.1 Monthly Plutonium Reports

A series of "Monthly Plutonium Reports" covering the period 1953 through 1961 were found in retired records located in Building K-1034,.[22] Labeling of the file indicates that these specific reports were discontinued in 1961. The reports contain results of health physics air monitoring and surface wipe samples and evidence a routine monitoring program for uranium and Pu. Sample descriptions identify the building, location, and operation sampled and include information about the source of the material being processed at the time of the sample (e.g., Savannah River oxide and Hanford oxide). Data contained in the reports were compiled into a data set. The data set contains results of Pu and uranium analysis for 298 samples taken in buildings K-1131, K-1231, K-132, K-1413 and K-1004-J. No Pu was detected in 165 of the 298 samples, and Pu ranged from 3 ppb-U to 18,833 ppb-U for the remaining 135 samples. Table 4.6.1-1 shows the 20 samples with the highest Pu results.

[22] A. F. Becher to J. Dykstra, "Monthly Plutonium Report," Union Carbide Internal Correspondence, January 1953 through July 14, 1961.

Table 4.6.1-1. Health Physics Monitoring Results - 20 Highest Pu Results

Year	Bldg	Desc. of Operation	Location	U *(mg)*	U *(c/m/ft3)*	Pu *(mg)*	Pu *(c/m/ft3)*	Pu *(ppb U)*
1957	K-1131	Routine Operations	Cold Trap Area, Center of UF6 Pumps	0.0060	0.0090	1.13E-07	0.0200	18,833
1956	K-1131	Routine Operations	Cold Trap Area Near Barrier Filters	0.0200	0.0290	1.80E-07	0.0290	9,000
1953	K-1131	Hydrofluorination and fluorination of Uranium compounds to furnish process material and removal	VH #1	0.0790	0.2000	5.00E-07	0.1150	6,329
1959	K-1131	Routine UF6 manufacture	"B" Line, East End of Reactor	0.0200	0.0300	6.45E-08	0.0100	3,224
1954	K-1131	Towers not operating; UF6 header open on tower platform	35' Tray Area	0.0063	0.2100	1.60E-08	0.0036	2,540
1954	K-1131	Ash Receiver Change and Dismantling of UF6 Lines and Towers	West End 35' Tray	0.1030	0.2470	2.15E-07	0.0480	2,087
1954	K-1231	Wipe samples were taken on top of the pulverizing unit and inside the material entry port. Unit was not in operation.	K-1231, Top of Pulverizing Unit	21.8800		3.64E-05		1,664
1953	K-1131	Hydrofluorination and fluorination of Uranium compounds to furnish process material and removal	Tower #1	0.2060	0.5020	3.20E-07	0.0736	1,553
1955	K-1131	Routine Operations	Barrier Traps	0.0300	0.0640	4.50E-08	0.0090	1,500
1955	K-1131	Routine Operations	Near Barrier Filters	0.0900	0.1570	1.29E-07	0.0210	1,433
1954	K-1131	Routine Tower and Tray Operation	Tower Platform	1.0900	2.7000	1.37E-06	0.3100	1,257
1954	K-1231	Wipe samples were taken on top of the pulverizing unit and inside the material entry port. Unit was not in operation.	K-1231, Top of Pulverizing Unit	17.3700		1.96E-05		1,128
1959	K-1131	Routine UF6 manufacture	"B" Line, East End of Trays	0.0600	0.1300	6.09E-08	0.0120	1,015
1954	K-1231	Wipe samples were taken on top of the pulverizing unit and inside the material entry port. Unit was not in operation.	K-1231, Top of Pulverizing Unit	33.8700		2.96E-05		874
1955	K-1131	Routine Operations	Tray Area	0.0200	0.0500	1.70E-08	0.0050	850
1957	K-1131	Routine Operations	South Wall of Screener Area	0.1400	0.2300	1.08E-07	0.0200	771
1957	K-1231	No Operations in Progress	Center of Pulverizing Area	0.0300	0.0400	2.10E-08	0.0020	700
1955	K-1231	Samples were taken during pulverizing operation.	Near Blender Booth	1.5600	11.0000	1.08E-06	0.6670	692
1957	K-1231	No Operations in Progress	Between Booths	0.0300	0.0300	2.00E-08	0.0020	667
1955	K-1131	Routine Operations	Cold Trap Area Near Barrier Trap	0.0400	0.0600	2.50E-08	0.0030	625

Table 4.6.1-2 shows the results of samples taken of the area in Building K-1004-J where laboratory analysis for Pu was performed. The data shown includes results of air samples as well as surface wipe samples from the work area. No Pu was detected in the samples.

Table 4.6.1-2. Health Physics Monitoring Results - Pu Sample Analysis Area

Year	Bldg	Desc. of Operation	Location	U *(mg)*	U *(c/m/ft 3)*	Pu *(mg)*	Pu *(c/m/ft 3)*	Pu *(ppb U)*
1956	K-1004-J	Routine plutonium analysis of uranium feed material and ash	2' from Hood	0.40	0.04	0.00	0.00	0.00
1956	K-1004-J	Routine plutonium analysis of uranium feed material and ash	2' from Hood "F"	0.50	0.03	0.00	0.00	0.00
1956	K-1004-J	Routine plutonium analysis of uranium feed material and ash	2' from Hood "G"	0.30	0.03	0.00	0.00	0.00
1956	K-1004-J	Routine plutonium analysis of uranium feed material and ash	Floor at Hood "G"	0.00	0.00	0.00	0.00	0.00
1956	K-1004-J	Routine plutonium analysis of uranium feed material and ash	Inside Hood "E"	0.00	0.00	0.00	0.00	0.00
1956	K-1004-J	Routine plutonium analysis of uranium feed material and ash	Inside Hood "F"	0.00	0.00	0.00	0.00	0.00
1956	K-1004-J	Routine plutonium analysis of uranium feed material and ash	Inside Hood "G"	0.00	0.00	0.00	0.00	0.00
1956	K-1004-J	Routine plutonium analysis of uranium feed material and ash	On Top of Sink	0.00	0.00	0.00	0.00	0.00
1956	K-1004-J	Routine plutonium analysis of uranium feed material and ash	Table in Center of Room	0.00	0.00	0.00	0.00	0.00
1956	K-1004-J	Routine plutonium analysis of uranium feed material and ash	Top of Funnel Rack	0.00	0.00	0.00	0.00	0.00

Table 4.6.1-3 shows the results of samples taken in Building K-132 during operations to unplug the UF_6 evacuation line used to “de-smoke” ash receivers. Table 4.6.1-4 shows results associated with changing barrier filters in Building K-1131. Table 4.6.1-5 shows results of air samples associated with the pulverizing operations in K-1231. Table 4.6.1-6 shows results of smear samples in and around the K-1231 ash pulverizing equipment.

Table 4.6.1-3. Health Physics Monitoring Results
K-132 Unplugging Operation

Year	Bldg	Desc. of Operation	Location	U (mg)	U (c/m/ft^3)	Pu (mg)	Pu (c/m/ft^3)	Pu (ppb U)
1954	K-132	Samples were taken during an unplugging operation of the UF_6 evacuation line used to "de-smoke" ash receivers.	Northeast Corner	2.01	11.28	0	0	0
1954	K-132	Samples were taken during an unplugging operation of the UF_6 evacuation line used to "de-smoke" ash receivers.	Platform, East Side	3.76	21.06	1.3E-07	0.067	35
1954	K-132	Samples were taken during an unplugging operation of the UF_6 evacuation line used to "de-smoke" ash receivers.	Platform, South Side	5.19	26.17	0	0	0

Table 4.6.1-4. Health Physics Monitoring Results
K-1131 Changing Barrier Filters

Year	Bldg	Desc. of Operation	Location	U (mg)	U (c/m/ft^3)	Pu (mg)	Pu (c/m/ft^3)	Pu (ppb U)
1953	K-1131	Changing Barrier Filters	North & South Barrier	1.33	32.00	5.8E-07	1.01	436
1953	K-1131	Changing Barrier Filters	North & South Barrier	3.61	14.60	1.1E-06	0.39	307
1953	K-1131	Changing Barrier Filters	North & South Barrier	21.62	72.60	1.4E-06	0.43	65
1953	K-1131	Changing Barrier Filters	North & South Barrier	3.86	19.46	5E-08	0.02	13
1953	K-1131	Changing Barrier Filters	Wipe 413A	87.30		1.3E-05		152
1953	K-1131	Changing Barrier Filters	Wipe 414A	45.00		9.2E-06		205
1953	K-1131	Changing Barrier Filters	Wipe 415A	313.00		3.1E-05		97

Table 4.6.1-5. Health Physics Monitoring Air Sample Results
K-1231 Pulverizing Operation

Year	Bldg	Desc. of Operation	Location	U (mg)	U (c/m/ft^3)	Pu (mg)	Pu (c/m/ft^3)	Pu (ppb U)
1955	K-1231	Samples were taken during pulverizing operation.	Near Blender Booth	1.56	11.00	1.1E-06	0.667	692
1955	K-1231	Samples were taken during pulverizing operation.	Near Pulverizer Booth	1.54	10.00	1.5E-07	0.093	98
1955	K-1231	Samples were taken during pulverizing operation.	Top of Pulverizer Booth	5.72	41.00	6.5E-07	0.429	113
1955	K-1231	Samples taken during the sampling and sealing of drums.	Near Blender Booth	0.15	0.49	0	0.000	0
1955	K-1231	Samples taken during pulverizing operations.	Inside Pulverizer Booth	85.62	205.00	8.7E-07	0.190	10
1955	K-1231	Samples taken during pulverizing operations.	Near Pulverizer Door	3.58	7.10	0	0.000	0
1955	K-1231	Samples taken during pulverizing operations.	Top of Pulverizer Platform	1.50	3.90	0	0.000	0
1956	K-1231	Pulverizing Operations	Center of Pulverizing Booth	4.61	10.32	4.4E-07	0.089	95
1956	K-1231	Pulverizing Operations	In Pulverizer Booth	136.00	381.00	4.1E-06	1.040	30
1956	K-1231	Pulverizing Operations	Platform Near Pulverizer	6.71	12.53	2.4E-06	0.407	356
1956	K-1231	Pulverizing Operations	Platform Near Pulverizer	2.80	5.90	1.1E-07	0.020	40
1954	K-1231	Pulverizing of ash - unit in operation during time of air sampling	10' North of Pulverizer Door	1.92	7.40	4.3E-07	0.154	224
1954	K-1231	Pulverizing of ash - unit in operation during time of air sampling	Pulverizer Platform	3.47	12.50	3.7E-07	0.121	105
1957	K-1231	No pulverizing operations in progress; grease seal on pulverizer being replaced by maintenance.	Center of North End	0.04	0.05	0	0.000	0
1957	K-1231	No pulverizing operations in progress; grease seal on pulverizer being replaced by maintenance.	Pulverizing Booth	0.04	0.05	0	0.000	0
1957	K-1231	No pulverizing operations in progress. Only normal or depleted uranium processed during the past 3 months.	Inside Blender Booth	0.11	0.15	0	0.000	0
1957	K-1231	No pulverizing operations in progress. Only normal or depleted uranium processed during the past 3 months.	Inside Pulverizing Booth	0.14	0.16	0	0.000	0
1957	K-1231	No Operations in Progress	Between Booths	0.03	0.03	2E-08	0.002	667
1957	K-1231	No Operations in Progress	Center of Pulverizing Area	0.03	0.04	2.1E-08	0.002	700
1954	K-1231		Between Pulverizer and Blender	0.43	1.27	8.5E-08	0.024	198

Table 4.6.1-6. Health Physics Monitoring Smear Sample Results
K-1231 Pulverizing Operation

Year	Bldg	Desc. of Operation	Location	U *(mg)*	Pu *(mg)*	Pu *(ppb U)*
1954	K-1231	Wipe samples were taken on top of the pulverizing unit and inside the material entry port. Unit was not in operation.	K-1231, Top of Pulverizing Unit	21.88	3.6E-05	1,664
1954	K-1231	Wipe samples were taken on top of the pulverizing unit and inside the material entry port. Unit was not in operation.	K-1231, Top of Pulverizing Unit	17.37	2E-05	1,128
1954	K-1231	Wipe samples were taken on top of the pulverizing unit and inside the material entry port. Unit was not in operation.	K-1231, Top of Pulverizing Unit	33.87	3E-05	874
1954	K-1231	Wipe samples were taken on top of the pulverizing unit and inside the material entry port. Unit was not in operation.	K-1231, Top of Pulverizing Unit	35.48	1.5E-05	428
1954	K-1231	Wipe samples were taken on top of the pulverizing unit and inside the material entry port. Unit was not in operation.	K-1231, Top of Pulverizing Unit	23.79	8.7E-06	365
1954	K-1231	Wipe samples were taken on top of the pulverizing unit and inside the material entry port. Unit was not in operation.	K-1231, Top of Pulverizing Unit	28.51	9.3E-06	325
1954	K-1231	Wipe samples were taken on top of the pulverizing unit and inside the material entry port. Unit was not in operation.	K-1231, Top of Pulverizing Unit	25.88	3.9E-06	150
1954	K-1231	Wipe samples were taken on top of the pulverizing unit and inside the material entry port. Unit was not in operation.	K-1231, Top of Pulverizing Unit	31.89	4.4E-06	138
1954	K-1231	Wipe samples were taken on top of the pulverizing unit and inside the material entry port. Unit was not in operation.	K-1231, Top of Pulverizing Unit	42.88	4.5E-06	105
1954	K-1231	Wipe samples were taken on top of the pulverizing unit and inside the material entry port. Unit was not in operation.	K-1231, Top of Pulverizing Unit	55.06	1E-06	19
1955	K-1231	Unit not in operation. Samples reported are surface smear samples.	Floor of Booth	1.86	6.8E-08	37
1955	K-1231	Unit not in operation. Samples reported are surface smear samples.	Floor of Main Room	0.53	3.9E-08	73
1955	K-1231	Unit not in operation. Samples reported are surface smear samples.	Floor of Platform	22.00	1.5E-07	7
1955	K-1231	Unit not in operation. Samples reported are surface smear samples.	Mouth of Hopper	24.50	3.4E-06	138

4.6.2 Fluorination of Special Nuclear Materials in K-1420

Information about the November 1960 processing of two shipments of HEU uranium oxide received from Hanford and Savannah River was found in retired records located in Building K-1034. The information consisted of correspondence, in the form of a letter report, from the Safety and Health Physics Organization to Operations.[23] The report stated that the material was processed through the K-1420 fluorination tower in two runs made November 3-9 and 9-14, 1960. The letter states that processing of the Savannah River Operations (SRO) shipment was of special interest because of the

[23] A. F. Becher to J. Dykstra, "Fluorination of Special Nuclear Materials—K-1420, November 3-14, 1960," Union Carbide Internal Correspondence, January 26, 1961

possible concentration of fission products in the system. Processing of the Hanford shipment was notable because the system had not been used previously to fluorinate uranium at "these high enrichments." Examination of the two sets of material yielded the following information:

- Beta-gamma survey of 15 process system locations (for Savannah River material).
- Spot-air samples of 15 locations at or near the process area (for the Hanford material) to delineate the area affected by the operation. The results are shown in Table 4.6.2-1.
- Continuous air monitor (CAM) results from t wo CAMs: one located adjacent to the west end of cold trap F-20-B in "E" area, and one located adjacent to the system charging point in "H" area. Data reported are shift averages and peaks and are shown for the two material runs in Table 4.6.2-2.
- Special bioassay of affected Maintenance and Operations personnel (20 workers).

Table 4.6.2-1. Spot-Air Sample Results from K-1420 Fluorination Tower Process November 1960

Spot-Air Sample Results - Hanford Material		
Location	**Start of Operations Alpha Activity ($c/m/ft^3$)**	**During Operation Alpha Activity ($c/m/ft^3$)**
H Area - 6' North of Tower	8.0	993.000
H Area - 2' from Open Can		52.000
E Area - B Cold Trap	2.0	0.400
E Area - Tower Room	25.0	10.000
B Area - 16' from West Wall		0.840
C Area - Column D-6		1.000
Outside E Area West Wall		0.210
E Area - Elevator Door		0.300
C Area - Column G-6		1.180
Outside E Area East Wall		0.240
E Area - Stairwell		2.520
C Area - Column G-10		0.320
Outside E Area North Wall Near Exhaust Fan		0.330
C Area - Column G-2		0.400
Roof - Near E Area Vent		0.100
Roof - Near H Area Vent		0.200
E Area Tower Room		13.800
F Area Center		1.500

- Additional gamma and alpha checks on samples from routine, off-site environmental air surveys that had shown higher than normal results for the period coincident with the two runs.
- Sampling of K-1420 roof surfaces and measurements and analysis for alpha counts, uranium, and ^{235}U.

Table 4.6.2-2. Shift Averages from Continuous Air Monitors During Processing of Savannah River and Hanford Material

Date	12-8 Shift Activity ($c/m/ft^3$)		8-4 Shift Activity ($c/m/ft^3$)		4-12 Shift Activity ($c/m/ft^3$)	
	Average	Peak	Average	Peak	Average	Peak
Savannah River Material						
"H" Area						
11/09/60			977.90	11,397.12	125.44	1,296.64
11/10/60	26.88	87.00	32.00	97.28	812.80	7,984.64
11/11/60	550.40	4.76	972.80	6,475.52	2,257.92	5,459.20
11/12/60	1401.60	4,066.56	1,491.20	7,240.96	32.12	299.52
11/13/60	666.88	6,560.00	64.0	803.84	440.32	4,616.96
11/14/60	19.20	140.80	116.48	1,126.40	64.00	787.20
11/15/60	12.80	1,437.44	75.52	186.88	592.64	6,856.96
11/16/60	131.84	42.24	125.44	893.44	44.80	241.92
11/17/60	24.32	51.20	17.92	58.88	21.76	186.88
11/18/60	7.68	15.36	15.36	46.08	35.84	240.64
11/19/60	26.88	112.64	14.08	53.76	6.40	47.36
11/20/60	7.68	17.92	398.08	4,268.80	72.96	487.28
11/21/60	16.64	42.24	47.36	3,146.88	20.48	103.68
11/22/60	3.84	5.12	6.40	39.68	69.12	262.40
11/23/60	8.96	88.40				
"E" Area						
11/09/60			8.55	49.14	1.61	4.67
11/10/60	2.77	10.79	4.60	44.67	5.91	21.13
11/11/60	2.48	16.55	15.01	50.63	6.80	19.54
11/12/60	14.40	46.57	19.62	29.29	11.39	44.47
11/13/60	13.16	46.14	2.76	25.89	9.75	41.34
11/14/60	0.79	1.83	4.17	39.61	6.51	36.69
11/15/60	0.59	0.72	3.52	14.78	4.63	2.50
11/16/60						
11/17/60	2.60		0.67	2.29	0.67	1.44
11/18/60	2.73	7.05	0.45	0.86	0.71	3.45
11/19/60	11.09	41.40	4.56	39.28	0.36	0.61
11/20/60	0.50	0.61	3.77	39.82	13.60	43.13
11/21/60	7.00	39.68	0.77	2.48	5.72	27.51
11/22/60	7.13	43.34	6.58	21.31	1.44	4.14
11/23/60	4.33	45.53	6.58	21.31	1.44	4.14
Hanford Material						
"H" Area						
11/04/60	0.55		(1) 4427.50	6,515.20	(2) 1624.30	11,032.30
11/05/60	638.70	5,899.50	16.60	38.40	147.20	590.00
11/06/60	17.90	88.30	1,358.00	10,695.70	472.30	3,576.30
11/07/60	12.80	29.40	106.20	331.50	165.00	769.30
11/08/60	47.40	327.70	271.40	1,272.00	650.20	5,017.60
11/09/60	1,857.30	14,720.00				
"E" Area						
11/04/60	6.40	37.50	9.41	43.89	2.42	8.69
11/05/60	8.07	42.20	11.11	9.96	0.77	2.62
11/06/60	1.35	2.90	8.27	25.67	11.51	45.49
11/07/60	0.43	0.87	8.31	22.46	10.61	37.71
11/08/60	2.85	27.77	8.56	37.17	10.65	46.84
11/09/60	8.22	47.06				

(1) Feed Screw Broken. Feed Hopper emptied pneumatically. Feed screw removed.
(2) Feed Hopper leaked. New gasket installed. Hopper charged.

The letter states that soon after start-up of the first run it became apparent that the air activity levels would be considerably in excess of the PAL[24], with average levels of 6.62 c/m/ft^3 and 787.2 c/m/ft^3 obtained for the "E" and "H" Areas respectively. The primary source of air-borne contamination was reported as being in "H" area, involving the pulverizer, hopper, and feed screw units. Peak periods were associated with operations of system opening, pneumatic transfer of material, and maintenance of equipment. The letter also notes that during the first four days of the operations, wearing of respirators by the personnel assigned was poor. Results of urinalysis for 11 of the 20 affected personnel showed uranium alpha counts in excess of the established control values, and those 11 were scheduled for recall visits. Excretion rates of all of the employees involved subsequently dropped below the follow-up level. Based on the information obtained by the evaluation, additional engineering controls (filtered containment enclosure) were recommended for the screw feed and hopper units.

4.6.3 Assessment of Accessible Contamination at the K-25 Site

In 1990, sampling conducted at PGDP suggested that levels of TRU contaminants at PGDP might be higher than previously estimated. A phased assessment program was undertaken at the GDPs, including the K-25 Site, to determine the magnitude of non-uranium radionuclides present in contamination in the process areas of the plants. The purpose of the assessment was to evaluate the potential impact of non-uranium radionuclides on the internal exposure control programs at the site. Results of that assessment are reported in *Assessment of Accessible Contamination at the K-25 Site Phase 3 Report: Cumulative Analytical Results*, May 1994.[25] Useful information provided by that report included the following:

- K-25 Pu urinalysis records (indicating negative results) exist from the early 1950's.
- In 1977, special air samples were analyzed for Np, Pu, and Am and controls were instituted based on the Np and ^{99}Tc results.
- For the assessment, samples were collected from a broad cross-section of the areas where contamination was exposed during the study. These samples are viewed as likely to be representative of the materials one would encounter during everyday activities within the site buildings where they were collected.
- The assessment was based on 96 samples taken in 19 buildings. Only one gamma-emitting fission or activation product, Cs-137, was reliably detected in contamination samples at the K-25 Site. It was found in only three samples.

Analysis methods included gamma ray spectroscopy for fission and activation products and separative chemistry followed by appropriate counting for ^{99}Tc, uranium isotopes, and transuranic radionuclides. Quality assurance aspects of the analysis are documented in the assessment report.

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[24] The PAL acronym was seen defined alternatively as Plant Acceptance Limit, Plant Allowable Limit and Plant Action Limit. The correct definition and usage remains to be confirmed.

[25] *Assessment of Accessible Contamination at the K-25 Site Phase 3 Report: Cumulative Analytical Results*, K/HS-570, Oak Ridge K-25 Site, May 1994.

Fig. 4.6.3-1 presents results reported for the ratio of ^{99}Tc to U activity in samples collected from buildings where contamination was exposed. In Fig. 4.6-1, the ^{99}Tc to U ratio was plotted by building, and the buildings were ordered approximately in accordance with the order material was processed through the ORGDP. Fig. 4.6.3-2 shows a plot of TRU to U ratios and was constructed in the same manner as Fig. 4.6.3-1.

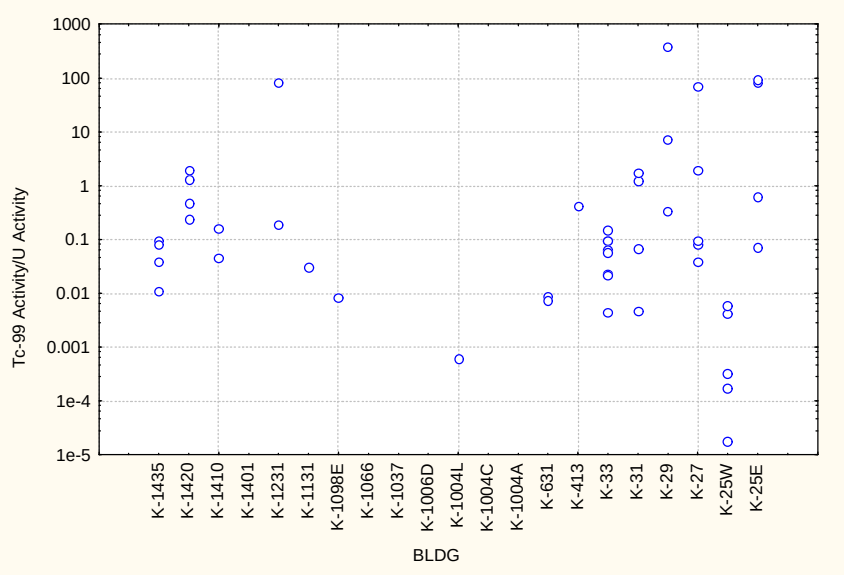

Fig. 4.6.3-1. Ratio of ^{99}Tc to U Activity in Accessible Contamination Samples.

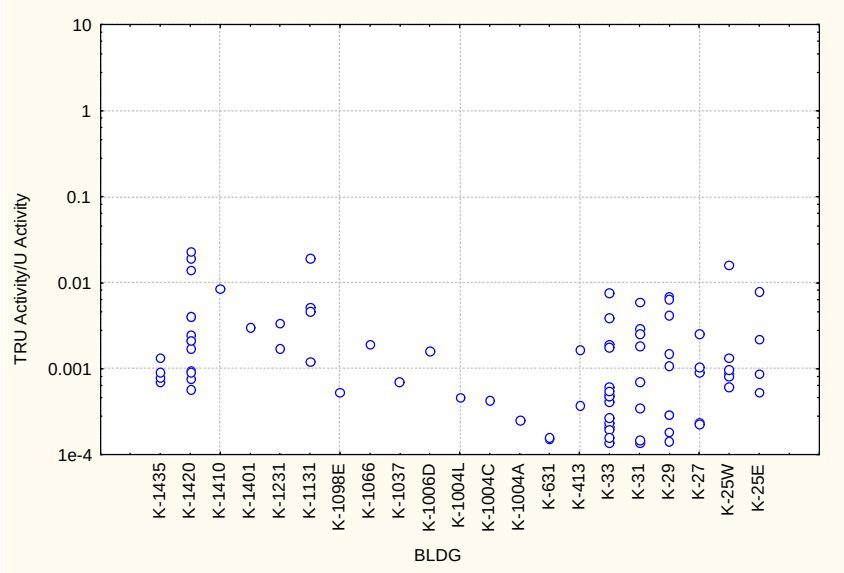

Fig. 4.6.3-2. Ratio of TRU to U Activity in Accessible Contamination Samples.

Recent safety authorization basis documentation[26, 27] indicates that large areas of the LEU and HEU buildings have radioactive surface contamination. Selected areas of Buildings K-33, K-31, K-29, K-25, and K-27 are designated as High Contamination Areas because of ^{99}Tc contamination. These areas are the north side of the Building K-33 operating floor, the west end of the Building K-31 operating floor, the entire cell floor of Building K-29, the north and south ends of the east side of K-25 on the cell and operating floors, in K-27 on the east and west ends of the cell floor, and in many cell areas on the K-27 operating floor (covering approximately 50% of the floor area). High Contamination is defined as activity levels exceeding 10,000 dpm/100 cm^2 from removable contamination and 50,000 dpm/100 cm^2 from fixed and removable contamination. In general, the ^{99}Tc contamination is characterized as easily removable. The characterization of the Contamination Areas is based on the results of radiological work permit job specific surveys and the large area wipe survey completed in 1994.

4.7 ANALYTICAL RESULTS FOR TRU AND FISSION PRODUCTS IN MATERIAL RELEASES ASSOCIATED WITH RU AT ORGDP

The *Oak Ridge Gaseous Diffusion Plant Historical Uranium and Radionuclide Release Report*[28] serves as the most comprehensive source found for information regarding material releases. This report compiled available historical data on the quantities of uranium and various radionuclides (including Pu, Np and ^{99}Tc) released from ORGDP from 1946 through 1984. The historical release data are organized into three major categories: airborne releases, liquid effluent releases, and on-site solid waste burial. The report contained no data indicating burial of material containing Pu, Np, or ^{99}Tc.

Because of the historical uranium accounting requirements at ORGDP, uranium release data are fairly extensive. However, the data for the other radionuclides are limited. Intermittent data were used, as appropriate, to tabulate quantities of radionuclides released. No attempt was made to extrapolate data for those years in which data were not available. Radiation levels are expressed in curies to depict release totals. Because the same quantity of each radionuclide generates a different level of radioactivity, the curie was used to standardize measurements of radioactivity released and to allow comparisons to be made.

4.7.1 Airborne Emissions

The *Oak Ridge Gaseous Diffusion Plant Historical Uranium and Radionuclide Release Report* indicates that the primary source of uranium and ^{99}Tc emissions have

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[26] *Basis for Interim Operation of the Low-enriched Uranium (LEU) Process Buildings at the East Tennessee Technology Park (ETTP)*, K/OPS-038, Rev.1, October 1997.
[27] *Basis for Interim Operation of the High-Enriched Uranium (HEU) Process Buildings at the K-25 Site*, K/OPS-050, Rev. 0, August 25, 1995.
[28] A. C. Lay and J. G. Rogers, *Oak Ridge Gaseous Diffusion Plant Historical Uranium and Radionuclide Release Report*, K/HS-95, ORGDP, February 28, 1986.

been the ORGDP purge cascade, the K-1131 feed plant, and several accidental releases. Airborne uranium emissions are presented in Table 4.7-1.

It is known that the feed plant was a major source of uranium air emissions during its operation, and ^{99}Tc was undoubtedly emitted. However, no data are available on the amount of ^{99}Tc emitted from the feed plant. The primary source of ^{99}Tc airborne emissions for ORGDP was the purge cascade. Beginning in 1974, the purge cascade vent was sampled continuously and analyzed on a daily and weekly basis. The data from those analyses were used to determine the ^{99}Tc emissions from ORGDP and are shown in Table 4.7-2. However, because the period of RU receipts from primary sources was 1952 to 1963, the greatest potential for ^{99}Tc emissions would have occurred during the period of 1953 to 1973 before this information was recorded.

Improved emission control equipment was installed on the purge cascade in 1977 in the form of solid chemical traps and a liquid potassium hydroxide scrubber. These improvements resulted in fewer emissions, as reflected in the data.

Uranium recovery processes were used in the K-1420 decontamination facility to avoid disposal of concentrated uranium solutions. However, residual concentrations of uranium, ^{99}Tc, Np, and Pu were released through liquid discharges. Liquid wastes discharged from the recovery operation were passed through K-1407-B and –C settling ponds where insoluble uranium compounds were removed and retained on-site. Soluble radionuclide compounds were discharged to Poplar Creek, which flows into the Clinch River.

Table 4.7-1. ORGDP Airborne Uranium Emissions 1946 - 1984

Year	Total Uranium Released *(curies)*	Total Uranium Released *(kg)*
1946	0.01	1
1947	<0.01	<1
1948	<0.01	5
1949	<0.01	45
1950	0.10	136
1951	0.02	146
1952	0.23	345
1953	1.60	*1,307
1954	0.26	68
1955	0.26	264
1956	0.81	225
1957	0.15	306
1958	1.80	*2,711
1959	1.10	531
1960	1.50	977
1961	3.10	773
1962	0.24	29
1963	3.10	*1,005
1964	0.01	7
1965	0.14	269
1966	<0.01	**1
1967	<0.01	2
1968	<0.01	<1
1969	<0.01	9
1970	<0.01	8
1971	0.02	21
1972	0.03	49
1973	0.13	144
1974	0.44	622
1975	0.27	371
1976	0.05	45
1977	0.03	17
1978	0.02	19
1979	0.04	25
1980	0.03	21
1981	0.01	5
1982	<0.01	2
1983	<0.01	2
1984	<0.01	1
Total	***15.61	***10,516

Note: The ratio between curies and mass differs from year to year due to varying isotopic enrichments.

* A major portion of the quantities reported in 1953, 1958, and 1963 resulted from accidental releases due to valve and trap failures in the K-402-1, K-1131, and K-1420 feed and processing facilities.

** Declining production levels was a factor, which reduced emissions in the 1966 to 1970 time period.

*** This total includes the actual stated value for any quantity which was reported as a less than (<) value.

Table 4.7-2. ORGDP Airborne ^{99}Tc Emissions 1974 - 1984

Year	^{99}Tc Released *(Curies)*	*^{99}Tc Released *(grams)*
1974	0.27	16
1975	0.3	18
1976	** 6.79	405
1977	***0.00	0
1978	0.29	17
1979	1.34	80
1980	0.88	53
1981	0.04	2
1982	0.03	2
1983	0.02	1
1984	0.02	1
Total	9.98	595

* This is based a ^{99}Tc activity of 59.7 g/Ci.
** This elevated valued may be due to increased purging of the cascade associated with the beginning of a large equipment changeout program that began in 1976.
*** This year the purge cascade location was changed from the K-25 Building to the K-29 Building. Data for both locations were added; however, the total amount was $2x10^{-6}$ curies/yr.

4.7.2 Liquid Emissions

The major radionuclides present in the liquid effluent were uranium and ^{99}Tc. Traces of Np and Pu were also present. ^{99}Tc liquid effluent releases are shown in Table 4.7-3. Np releases are shown in Table 4.7-4. This table shows that Np was found only in small quantities. Sampling of surface waters for Pu near the effluent of the uranium recovery operation revealed the presence of Pu only twice. On both occasions, the concentration was just above the detection limit of $0.1x10^{-14}$ ci/ml.

Table 4.7-3. Tc-99 in ORGDP Liquid Effluent

Year	^{99}Tc Released *(curies)*	^{99}Tc Released *(grams)*
1974	3.5	208
1975	9.0	539
1976	*24.1	1,437
1977	5.8	344
1978	4.0	239
1979	7.3	436
1980	5.1	307
1981	3.5	211
1982	1.7	100
1983	**17.0	1,018
1984	**10.1	604
Total	91.1	5,443

* This evaluated value may be due to increased decontamination efforts associated with the beginning of a large equipment change out program.
** In 1983 and 1984, there was a great amount of decontamination work performed on equipment from an area of the cascade highly contaminated with Tc-99. Also in 1983, there occurred a larger than normal technecium-99 release from the decontamination facility. The cause of this release was never determined.

Table 4.7-4. Np in ORGDP Liquid Effluent

Year	Np Released *(curies)*	Np Released *(grams)*
1979	0.0015	0.2
1980	0.0014	0.2
1981	0.0021	0.3
1982	0.0019	0.3
1983	0.0004	0.0
Total	0.0073	1.0

4.8 ANALYTICAL RESULTS FOR TRU AND FISSION PRODUCTS IN URANIUM MATERIALS SHIPPED FROM ORGDP

Results of TRU and fission product analyses performed on samples of ORGDP enriched product for the period 1983 through 1985 were found in a series of reports of natural and reactor return feed analyses.[29, 30, 31, 32] The reports summarized results of sampling and analysis performed at ORGDP for defining adherence to feed specifications. Summarized results for ORGDP product were included for comparison purposes. Product results were reported for shipments to both domestic and foreign fuel fabricators or enrichment customers. The number of analyses performed for ORGDP product was not as extensive as that performed for the foreign reactor returns feed to ORGDP. However, some analysis for TRU and fission products in ORGDP product was performed. The summarized results are shown in Table 4.8. None of the product samples analyzed exceeded specifications.

[29] W. D. Hedge, *Toll Enrichment Uranium Hexafluoride: Natural and Reactor Return Feed Analyses at ORGDP for CY 1982, Including Summaries for CYS 1969-1982*, K/TL/AT-58, Rev. 1 Addendum 2, Union Carbide Corporation Nuclear Division, April 1983.
[30] W. D. Hedge, *Analyses of ORGDP Toll Enrichment Uranium Hexafluoride for CY 1983*, K/PS-5034, Union Carbide Corporation, March 1984.
[31] W. D. Hedge, *Analyses of ORGDP Toll Enrichment Uranium Hexafluoride for CY 1984*, K/PS-5034, Addendum 1, Union Carbide Corporation, May 1985.
[32] W. D. Hedge, *Analyses of ORGDP Toll Enrichment Uranium Hexafluoride for CY 1985 Through September 1986*, K/PS-5034, Addendum 2, Union Carbide Corporation, January 1987.

Table 4.8. Summarized Results for TRU and Fission Products in ORGDP Enriched Uranium Product

CY	Property	U-235 *(wt%)*	U-236 *(wt%)*	Fission Product Gamma *(% Aged Natural U)*	Fission Product Beta *(% Aged Natural U)*	TRU Alpha *(dpm/gU)*	Tc *(ppm/U)*
1982	ORGDP Product	2.96900	0.0079	8.2	0.9	9.7	0.036
1983	ORGDP Stockpile	3.10144	0.0175	<0.1	<5.0	5.8	0.014
1983	French (Cogema)	2.88520	0.0050			<5	
1984	British (BNFL)	2.56632	0.0176	<0.1	1.1		0.279
1984	Japan	3.11195	0.0175	<0.1	0.8		
1984	Westinghouse Electric	3.26739	0.0137	<0.1	<5.0		0.056
1984	ORGDP Stockpile	3.06811	0.0207	<0.1	1.2		0.026
1985	ORGDP Stockpile	2.58587	0.0479	<5.0	1.5	3.5	0.014
1985	Japan	2.95407	0.0115	<5.0	<0.1	<5	0.028
1985	Westinghouse Electric	3.14285	0.0136	<5.0	<0.1	<5	0.046